



EFFECTS OF RE-EMISSION ON THE EVOLUTION OF
SURFACE MORPHOLOGY DURING THERMAL
EVAPORATION DEPOSITION

M.S. THESIS

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Submitted to the faculty of the Applied Science Department
University of Arkansas at Little Rock

Nov 9, 2010



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2010

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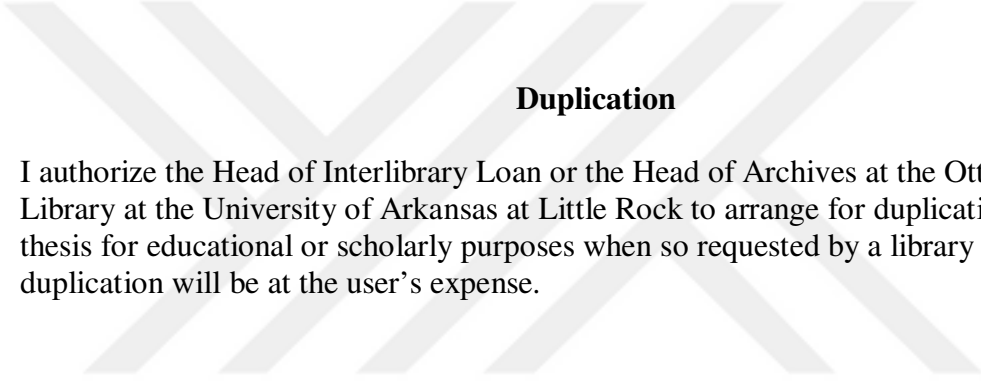
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EFFECTS OF RE-EMISSION ON THE EVOLUTION OF SURFACE MORPHOLOGY
DURING THERMAL EVAPORATION DEPOSITION by Mehmet Erdem, 2010

ABSTRACT

In re-emission process during thin film growth, atoms that do not stick to the surface based on a sticking probability are re-emitted and can stick to other surface points. This process allows atoms to reach valleys of the surface, which are normally shadowed by higher surface points, leading to a smoothening effect. Although re-emission has been applied to sputter deposition and chemical vapor deposition (CVD) growth modeling, which successfully predicts the dynamic morphological evolution, so far, no re-emission based modeling method has been developed for thermal evaporation growth. Thermal evaporation is a commonly used thin film deposition technique especially for low melting point elemental materials. Therefore, we developed a new growth modeling approach for thermal evaporation thin film deposition process that takes into the effects of re-emission. The modeling method is based on Monte Carlo simulation techniques and numerical solution of a continuum growth equation that include the effects of re-emission, shadowing, surface diffusion, and noise. Thin film morphologies obtained from Monte Carlo simulations and by numerically solving the growth equation were investigated for their dynamic evolution of surface roughness.

This study is dedicated to my wife, Yasemin, and my son, Baris.



Acknowledgements

I am very thankful to my advisor, Dr. Tansel Karabacak, who spent a lot of his precious time with me. I would also like to give thank to my committee members; Dr. Huang, Dr. Seo and Dr. Cui. In addition, I am also thankful for my friends Mr. Mehmet Fatih Cansizoglu and Mr. Arif Sinan Alagoz because of their helps in my training for experimental systems in our lab. I would like to give thanks to Dr. Abhijit Bhattacharya since he leaded us to learn refworks, which facilitated the writing style of references, and Brent Nelson for his precious explanations about the same program. Due to their time in training me for the Scanning Electron Microscopy (SEM) and Atomic Force Microscopy (AFM) systems, I am also very grateful for Dr. Fumiya Watanabe and staff of Nanotechnology Center at UALR.

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1. PLAN OF THESIS

Below, we outline the content of this thesis:

In chapter 2, we present background information about what thin film is. First, we explain what thin film deposition is. Second, we mention about surface roughness and explain important surface roughness parameters. Then, we briefly explain types of thin film surface growth models which are continuum models and discrete models. Since we study thermal evaporation deposition, we briefly explain thermal evaporation deposition as a subsection of this introductory chapter.

In chapter 3, we survey literature about thin film deposition techniques including sputter deposition, chemical vapor deposition and especially thermal evaporation deposition techniques. Although these studies are usually experimental, they also include theoretical results. We obtain data using Monte Carlo simulation methods and numerical solution methods; we also do literature survey about Monte Carlo simulations and numerical solution methods.

After giving the important terminology, we write our goals and objectives of the thesis in chapter 4.

Chapter 5 and 6 is one of the most important chapters throughout the thesis; we examine the effects of re-emission during thermal evaporation growth using Monte Carlo simulation methods with and without surface diffusion. We completely use flat substrate at the beginning. We explain what first and all order re-emissions are, and we compare the differences. We compare data and graphics with the following chapters since we use different parameters and techniques.

In chapter 7, all our parameters are the same except the initial substrate. We use patterned initial substrates and after evolution of the surface morphology, we compare the surface morphologies whether the initial substrate is flat or patterned.

Instead of using Monte Carlo simulation methods, we analyze the surface using numerical solution methods in chapter 8. We could not get enough data since numerical solutions are too slow.

Chapter 9 compares all of the data which we obtain so far. We summarize our work in this chapter and we present some recommendations for future work.

At the end of the thesis, we list the references that we use and plot the flow of the algorithms of Monte Carlo simulations and numerical solution methods that we use.

2. INTRODUCTION AND BACKGROUND

2.1 Thin Film Deposition

Thin films are coatings which range from fractions of a nanometer to several micrometers in thickness. They have a lot of applications in the semiconductor industry such as solar cells, micro-electro-mechanical systems (MEMS), nano-electro-mechanical systems (NEMS), computer chips, photonic bandgap materials, detectors, and sensors. They are all based primarily on the development/creation and utilization of different kinds of thin films which are deposition and patterning.

Thin film deposition is the act of applying a thin film to a surface. Among several thin film deposition techniques, two of them are commonly used for the growth of thin films with high material properties such as electrical, optical, and magnetic properties. One of these methods is chemical vapor deposition (CVD) and the other is physical vapor deposition. These types depend on whether the deposition process is mainly chemically or physically driven. “CVD is a chemical process used to produce high-purity, high-performance solid materials.”¹

In a typical CVD process, the wafer (substrate) is exposed to one or more volatile precursors², which react and/or decompose on the substrate surface to produce the desired deposit³. A number of forms of CVD are in wide use and are frequently referenced in the literature⁴. These processes differ in the means by which chemical reactions are initiated (e.g., activation process) and process conditions. Types of CVD are Atmospheric pressure CVD (APCVD)⁵, Low-pressure CVD (LPCVD)⁶, Ultrahigh vacuum CVD (UHVCVD)^{7,8}, Plasma-enhanced CVD (PECVD)⁹, Remote PECVD^{10,11}, Atomic layer

deposition (ALD)¹¹, Metalorganic chemical vapor deposition (MOCVD)¹², and Hybrid physical-chemical vapor deposition (HPCVD)¹³.

Another important vacuum deposition approach is physical vapor deposition (PVD), which is a general term used to describe the variety of methods to deposit thin films by the condensation of a vaporized form of the material onto various surfaces (e.g., onto semiconductor wafers)^{14, 15}. The coating method involves purely physical processes such as high temperature vacuum evaporation or plasma sputter bombardment rather than involving a chemical reaction at the surface to be coated as in CVD. Some of the well known types of PVD are electron-beam (e-beam)¹⁶, thermal evaporation¹⁷⁻²⁵, sputter deposition²⁶⁻²⁸, cathodic arc deposition²⁹⁻³¹, pulsed laser deposition³²⁻³⁴, and^{35, 36} resistive heating thermal evaporation^{35, 36}.

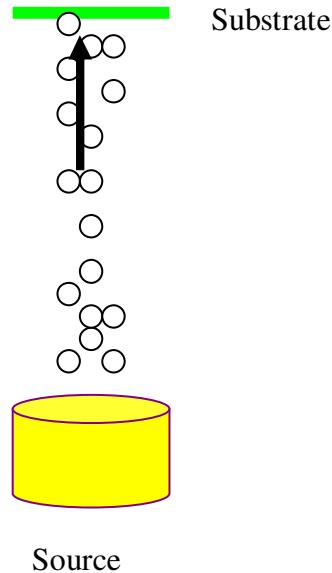


Figure 2.1: A schematic showing of a thermal evaporation deposition experiment.

Among many vacuum deposition methods, thermal evaporation deposition method is one of the simplest unlike such as sputter deposition that requires a plasma environment or CVD that involves complicated chemical processes. As illustrated in Fig. 2.1, the source material is placed in a crucible and then heated until it evaporates and condenses on a substrate located above the crucible³⁷. We will discuss thermal evaporation deposition in more detail in chapter 2.6. Thermal evaporation technique will be the choice of method of thin film deposition and the modeling for the thesis. In our modeling studies, we will investigate the important mechanisms that control surface evolution such as surface diffusion, re-emission, noise, and shadowing.

2.2 Surface Roughness

Surface roughness of thin films can affect many of the film properties such as its electrical conductivity, optical absorption, and friction. Therefore, characterization of surface roughness is an important task towards producing high quality thin films. For example, in semiconductor manufacturing, roughness of bare silicon wafers is measured after polishing and cleaning. Excessive roughness destroys the integrity of very thin layers, such as gate or tunnel oxide. Roughness is equally important for other electronic materials, such as SiC (silicon carbide) and GaAs (gallium arsenide)^{38,39}

In Fig. 2.2, some of the commonly employed deposition techniques are illustrated. The graph also shows a plot of the incidence flux distribution of atoms arriving at the substrate for these different deposition techniques. θ in Fig. 2.2. is the angle between the incident atomic flux and the surface normal. For thermal evaporation or e-beam evaporation, if the substrate is sufficiently far away from the source and if the substrate

dimensions are not too large, the flux arrives at the substrate with $\theta \sim 0^\circ$ which is referred to as normal incidence. This figure also shows commonly employed deposition techniques: thermal evaporation deposition, sputter deposition, chemical vapor deposition and oblique angle deposition. Last technique is also called glancing angle deposition (GLAD)⁴⁰⁻⁴⁵. “This technique is typically used for the growth of nanostructured arrays of rods and springs through a physical self assembly process.”⁴⁶

When the incident atoms come, they choose to land on taller islands. This process is named physical self assembly.⁴⁷ Presently, we do not include the molecular beam epitaxy (MBE). This plot also includes a graph showing the incident flux distribution for various deposition techniques.

“In many applications, it is often desired to have atomically flat thin film surfaces.”⁴⁶ However, in almost all of the deposition techniques mentioned above except MBE, the surface morphology generates a growth front roughness.⁴⁸ “The formation of growth front is a complex phenomenon and very often occurs far from equilibrium.”⁴⁶ “In order to measure the quantitative information about the morphology of a growth front, we use two classes of techniques. They are diffraction techniques and real-space imaging techniques. Undoubtedly, there are many examples for these two techniques. Examples for diffraction technique are high energy electron diffraction (RHEED), high resolution low-energy electron diffraction (HRLEED), atom diffraction, X-ray diffraction and light scattering. Examples for real-space imaging techniques are atomic force microscopy (AFM), scanning tunneling microscopy (STM), scanning electron microscopy (SEM) and stylus profilometry.

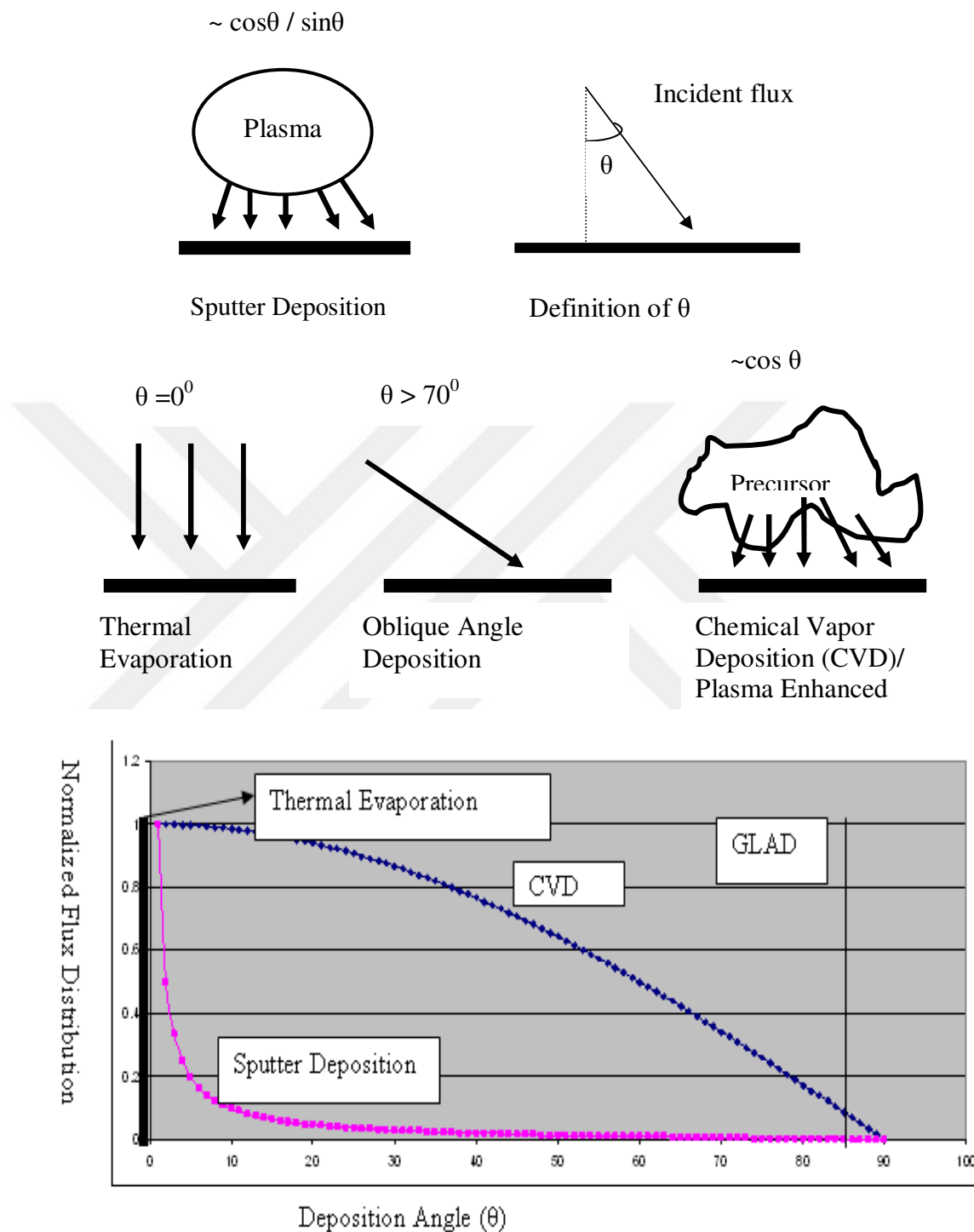


Figure 2.2: The geometries of several commonly employed deposition techniques. The graph is a plot of the incidence flux distribution of atoms arriving at the substrate for different deposition techniques.

This technique has the advantage of providing a direct visual interpretation of the surface morphology. However, the advantages of diffraction techniques include providing non-contact measurements and the ability to obtain a statistical average of a large surface area in a short time.”³⁷

There are statistical methods developed in order to define surface roughness in a quantitative way. One of the most frequently used ones is interface width ω , or also named root-mean-square (RMS) roughness. RMS roughness is defined as⁴⁹⁻⁵³

$$\omega(t) = \sqrt{\langle ([h(r,t) - h(\text{mean})]^2) \rangle} \dots \dots \dots (1)$$

where $h(\text{mean})$ is the average height, $h(r, t)$ is the height at a position ‘r’ and a time ‘t’. The mathematical definition between the symbols “<” and “>” tells us that we should take the statistical average.

Dynamic evolution of RMS roughness with growth time t generally obeys a power law behavior $\omega(t) \sim t^\beta$, where β is a growth exponent, and it is generally $0 \leq \beta \leq 1$.^{39, 54-59} Small β indicates that roughness evolution is slow and final surfaces are typically smooth, and large β indicates that roughness develops very quickly and final film morphology can be very rough.

We can measure β values from the relation below (in other words from the slope of log-log plot of ω versus t):

$$\log(\omega(t)) = \beta \log t + \text{const} \dots \dots \dots (2)$$

Another important roughness parameter is the lateral correlation length (ξ), which reflects the average lateral size of the “hills” on the thin film surface. Dynamic evolution of ξ also shows a power law behavior and it obeys $\xi \sim t^{1/z}$, where z is the dynamic

exponent.^{58, 60-62} Therefore, we can measure $1/z$ values from the slope of log-log plot of ξ versus t .



Figure 2.3: Illustration of large and small roughness exponents, α .

The next important roughness parameter is roughness exponent α , and it shows how wiggly the surface is and it is related to the fractal dimension of the surface.⁶³⁻⁶⁵ For example, see the illustration in Fig. 2.3. Both profiles in this figure have similar RMS roughness and lateral correlation values but the one at the bottom looks rougher in small scales. This small scale roughness can be defined through roughness exponent α . This exponent reflects how wiggly the surface is at length scales smaller than the lateral correlation length. α can be measured from a height-height correlation functions which is defined as^{58, 59, 61, 66, 67}

$$H(\mathbf{r}) = \langle [h(\mathbf{r}) - h(0)]^2 \rangle \dots\dots\dots(3)$$

where $H(r) \sim r^{2\alpha}$, $0 \leq \alpha \leq 1$. In height-height correlation function, we find the average values of squares of the difference between surface heights at a position, r , and at a reference height.

Even though surface heights are not significantly correlated, they may exhibit a periodic behavior on a length scale larger than the lateral correlation length. In order to determine this long range behavior the Power Spectral Density (PSD) function is used.^{51,}

^{61, 68} It is the Fourier transform of the surface heights.⁵³ Using this function, we can define whether a wavelength selection has or in other words whether we have a periodic behavior on the surface. λ , wavelength, which describes the separation of the mounds, can also change as a function of time in a power-law form $\lambda \sim t^p$ where p is the wavelength exponent.^{37, 68, 69} Now the question is how we can find whether there exists wavelength. One simple way of finding this out is through the analysis of PSD plots. If one cannot see a peak as in Fig. 2.4, we can say there is no wavelength selection. On the other hand, if there is, as illustrated in Fig. 2.5, a wavelength selection is described to exist. Peak position in PSD plots can be used to calculate wavelength λ values. For example, from the maximum peak frequency ($k_{\max}$) position in Fig. 2.5, we can calculate λ , by using $\lambda = 2\pi / k_{\max}$.

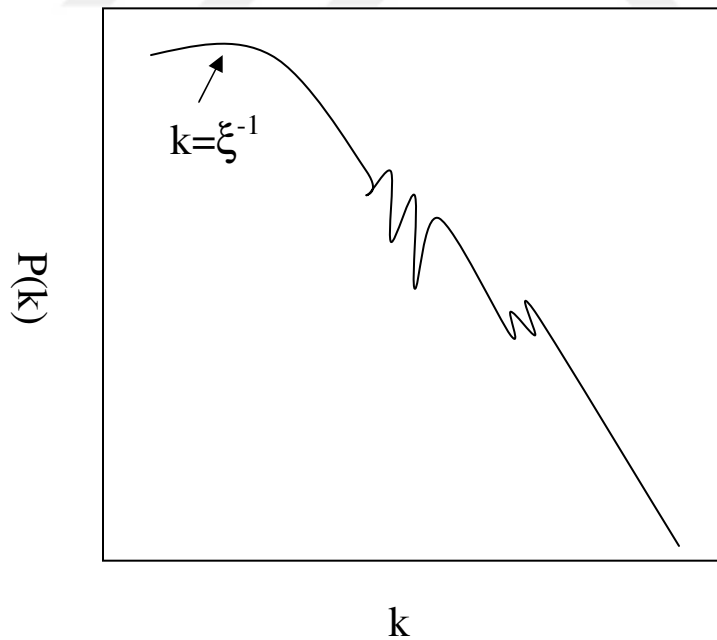


Figure 2.4: Illustration of power spectral density function that exhibits no characteristic peak.

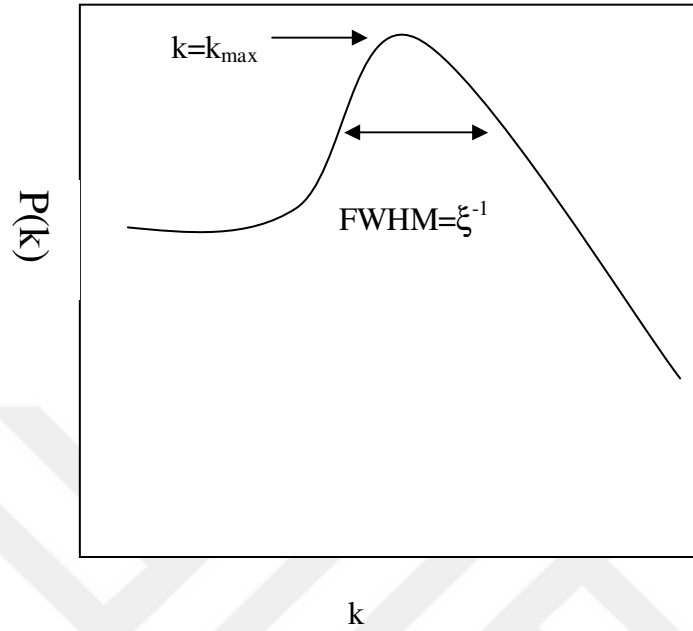


Figure 2.5: Illustration of power spectral density function that exhibits characteristic peak.

2.3 Important Mechanisms that Control Surface Roughness

There are many factors that contribute to the formation of a surface of a film. One of these factors is random noise.⁷⁰⁻⁷² “As atoms are deposited on surface, atoms do not arrive at the surface at the same time uniformly across the surface.”⁷³ Noise competes with the surface smoothening process. There is always random noise that exists naturally during growth, because atoms do not arrive at the surface uniformly. This reason may create surface growth front roughness.⁶¹

When an atom hits the surface, it moves around nearby atoms and then it sticks. This process is called surface diffusion.^{67, 74, 75}

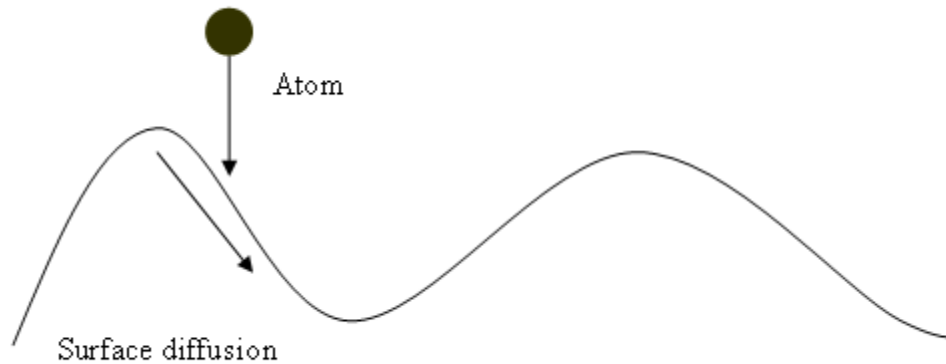


Figure 2.6: Illustration of surface diffusion.

Another important growth mechanism originates from that crest of the surface will receive more deposited atoms than the valley due to the shadowing effect^{51, 76} (see Fig. 2.7). Shadowing effect is normally long range or in other words non-local. Non-local shadowing competes with smoothening effects and wins at sufficiently long time. In many commonly deposition techniques such as sputter deposition and CVD, shadowing effect exist but its strength might vary depending on the detailed flux distribution profiles of incident atoms.

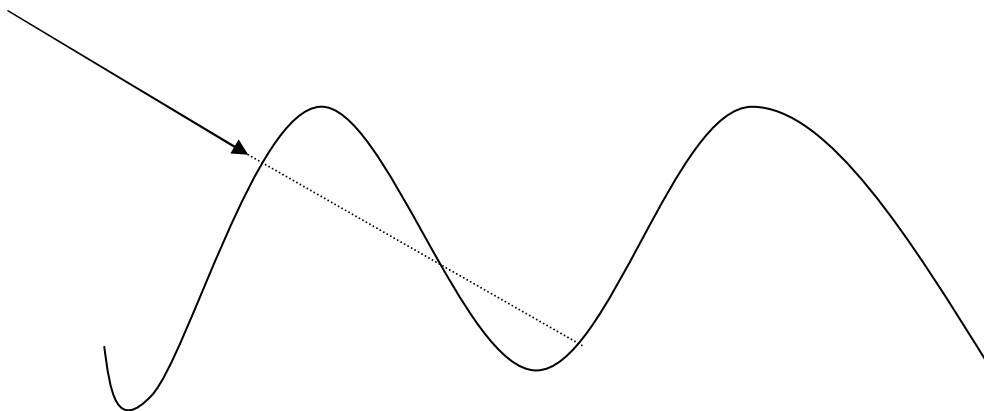


Figure 2.7: Illustration of the shadowing effect. The valley receives fewer particles than the peaks, because incoming particles are blocked by the peak.

In addition, when a particle comes and hits the surface, it either sticks or can leave the surface after several physisorption or chemisorption process. The average probability of an incident atom or molecule to stick to the surface is called sticking coefficient and denoted as S_c .^{50, 77, 78} If the particle doesn't stick, then it will be re-emitted (see Fig. 2.8) and go elsewhere. This process is called re-emission. If the particle re-emits and then sticks or goes elsewhere, we say that it is first order re-emission and if this process is repeated multiple times, we say that it is all order re-emission.^{51, 76, 78}

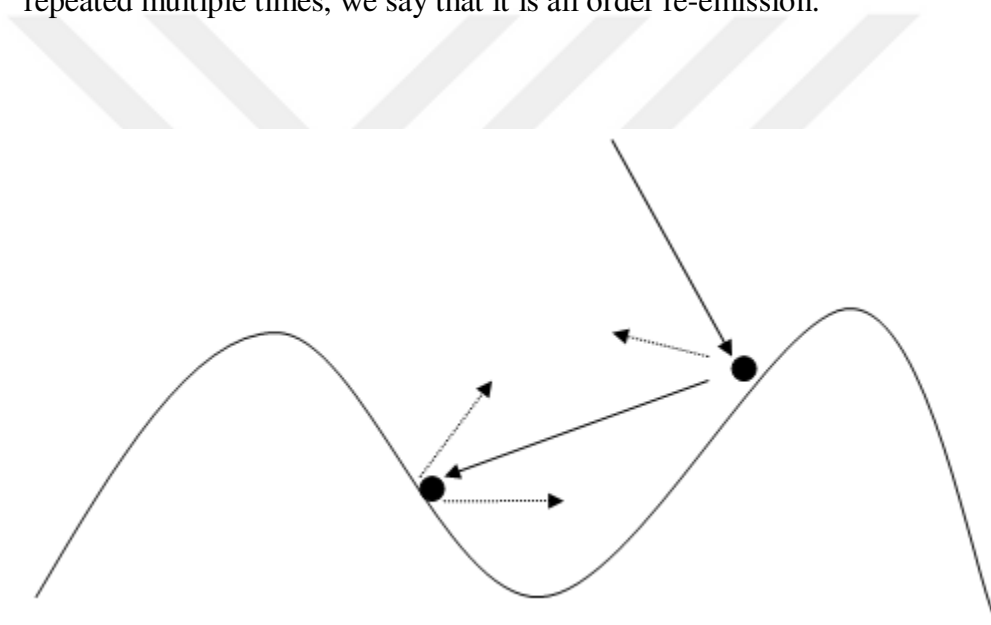


Figure 2.8: Illustration of re-emission effect.

2.4 Continuum Models

Continuum models of thin film growth are often expressed as partial differential equations. We can learn information about morphology by solving the continuum growth equation, which is

$$\frac{\partial h(\mathbf{r}, t)}{\partial t} = G + \eta(\mathbf{r}, t) \dots \dots \dots (4)$$

where G is the surface smoothing mechanisms and η is the random noise and can be described by^{37, 67, 68, 79}

$$\langle \eta(\mathbf{r}, t) \eta(\mathbf{r}', t') \rangle \sim \delta(\mathbf{r} - \mathbf{r}') \delta(t - t') \dots \dots \dots (5)$$

This type of model is not deterministic, and it is not possible to find an explicit solution for surface heights $h(\mathbf{r}, t)$. However, one can use it to predict average statistical properties of h such as the mean surface height and surface roughness.

The primary advantage of a continuous growth model lies in the ability to choose function G . Terms can be included in G depending on the type of growth effect one would like to model, which often come from considerations of how a certain growth effect would change the surface height profile $h(\mathbf{r}, t)$.

There are different versions of continuum equation all of which includes different mechanisms. These equations are;

Edward-Wilkinson (EW)⁶⁸

$$\frac{\partial h(\mathbf{r}, t)}{\partial t} = \nu \nabla^2 h + \eta(\mathbf{r}, t) \dots \dots \dots (6)$$

Here ν is called the surface tension, for the first term of the right side tends to smooth the interface. EW equation is the simplest equation describing the fluctuations of an equilibrium interface. This is the first continuum equation that is used to study the growth of interfaces by particle deposition. Solutions can be found by solving this equation: $\alpha = (2-d)/2$; $\beta = (2-d)/4$; and $z=2$. For $d > 2$, the roughness exponent α becomes negative and this means the interface is flat.⁶⁸

Kardar – Parisi - Zhang (KPZ)^{68, 80}

$$\frac{\partial h(\mathbf{r}, t)}{\partial t} = \nu \nabla^2 h + \lambda (\nabla h)^2 + \eta(\mathbf{r}, t) \dots \dots \dots (7)$$

When the predictions of the linear theory changed, nonlinear terms were added to the growth equation. KPZ is the first equation that includes nonlinear terms. Some exact results have been obtained, though KPZ equation cannot be solved in closed form because of its nonlinear character. The first term of the right hand side describes relaxation of the interface caused by a surface tension ν .

Wolf-Villain (WV) or Mullins equation⁶⁸

$$\frac{\partial h(\mathbf{r}, t)}{\partial t} = -\nu \nabla^4 h + \eta(\mathbf{r}, t) \dots\dots\dots(8)$$

The Wolf-Villain model was the first model introduced to investigate kinetic surface roughening in the absence of desorption in order to model dynamics of surfaces.⁷⁴

The first term of the right hand side is the surface diffusion term. This term has a smoothening effect. This is important in our study since we will do our study first assuming not including surface diffusion and then including surface diffusion. After that, we will compare our results and we are going to see the effect of re-emission. In this model the EW term, which is the second order of gradient, doesn't exist.

Lai, Das Sarma (LD)⁶⁸

$$\frac{\partial h(\mathbf{r}, t)}{\partial t} = \nu \nabla^2 h + \lambda (\nabla^2 \nabla h)^2 + \eta(\mathbf{r}, t) \dots\dots\dots(9)$$

This model introduces a geometrical construct to physically understand the features of various growth terms in the model for various growth terms. This enables us to identify a growth equation which may be relevant to the atomistic MBE process.⁷⁰

We are going to use WV since there is a surface diffusion term $(-\nabla^4 h)$. We can describe a universality which says that there are only distinct values of α , β , z .⁷² For

instance, according to Wolf-Villain, the prediction of α , β , and z are 1, $\frac{1}{4}$, 4 respectively where $z=\alpha/\beta$. Table 1 gives α , β , and z values of these four equations.^{37, 53, 68, 81}

	α	β	$z=\alpha/\beta$
Edward-Wilkinson	0	0	2
Kardar-Parisi-Zhang	0.38	~ 0.25	1.58
Wolf-Villain	1	0.25	4
Lai, Das Sarma	$\frac{2}{3}$	0.2	$\frac{10}{3}$

Table 2.1: α , β , and z values for four different models

These continuum models predict that independent of the materials being deposited or experimental details, one should get only a set of exponents as listed in Table 2.1 above. This concept is called “universality”. For example, if surface diffusion is dominant in a growth process then β is expected to be 0.25, $\alpha = 1$, and $z = 4$, as predicted by the Wolf-Villain (or Mullins) growth model, where it is believed the $-\nu \nabla^4 h$ term in this model represents the surface diffusion process.^{37, 46, 49, 53, 68}

2.5 Discrete Models

In addition to the continuum models, researchers also studied discrete growth models mainly through Monte Carlo (MC) simulation methods^{51, 56, 63, 76, 82, 83}, where the name Monte Carlo comes from the city which is famous for gambling. Like in gambling, MC simulations involve random events. For example, in a MC simulation used for thin film growth particles are sent to random positions on the substrate or during surface

diffusion atoms can hop toward random lateral positions. Why do people use Monte Carlo simulation methods instead of analytically or numerically solving continuum growth equations? As we will see in the following chapters, some advanced growth equations that can for example include re-emission and shadowing effects are almost impossible to solve analytically and numerical solutions requires very long computation times, which makes them unpractical. Other kind of simulation methods such as molecular dynamic (MD) simulations also requires high computational power, and yet it still can simulate only a handful of atoms for very limited growth times. However, we note that MD simulations involve atomistic potentials, are physically more realistic and can be utilized for example to predict sticking coefficient values to be later used in MC simulations as parameters inputs.³⁷

Unlike other methods, Monte Carlo simulations are fast and can simulate a large number of atoms. MC simulations do not involve atomistic details, however they can be used for the prediction of average morphological properties of thin film surfaces, such as RMS roughness, which is an average statistical property of the surface and do not strongly depend on atomistic details of the growth process. For example, MC simulations can use a sticking coefficient values as inputs to simulate the re-emission process, instead of simulating the process based on atomistic potentials, which requires enormous amount of computation power.³⁷

In MC simulations, the surface is assumed to be described by a height matrix $h(i, j)$ defined on a periodic $N \times N$ lattice. Furthermore, h can take on only integer values. The simulation proceeds one particle at a time. First, a random position in the x - y plane is chosen for the particle. The initial position of the particle in the h direction is always one

position higher than the maximum of the surface. Next, the direction that the particle will go off in is chosen; the distribution of directions depends on the properties of the incident flux. The particle moves in this direction until hitting the surface, after which it can be deposited on the surface or be re-emitted according to the re-emission mode. If it is re-emitted, the particle will travel until it hits the surface again or passes above the highest point on the surface. Once the particle has been deposited on the surface or has passed above the highest point on the surface (meaning it will never hit the surface), the above procedure is repeated for a new particle.^{55, 56, 61, 83, 84} By this way, MC simulations can accommodate important growth mechanisms and present a reliable way of growth modeling.

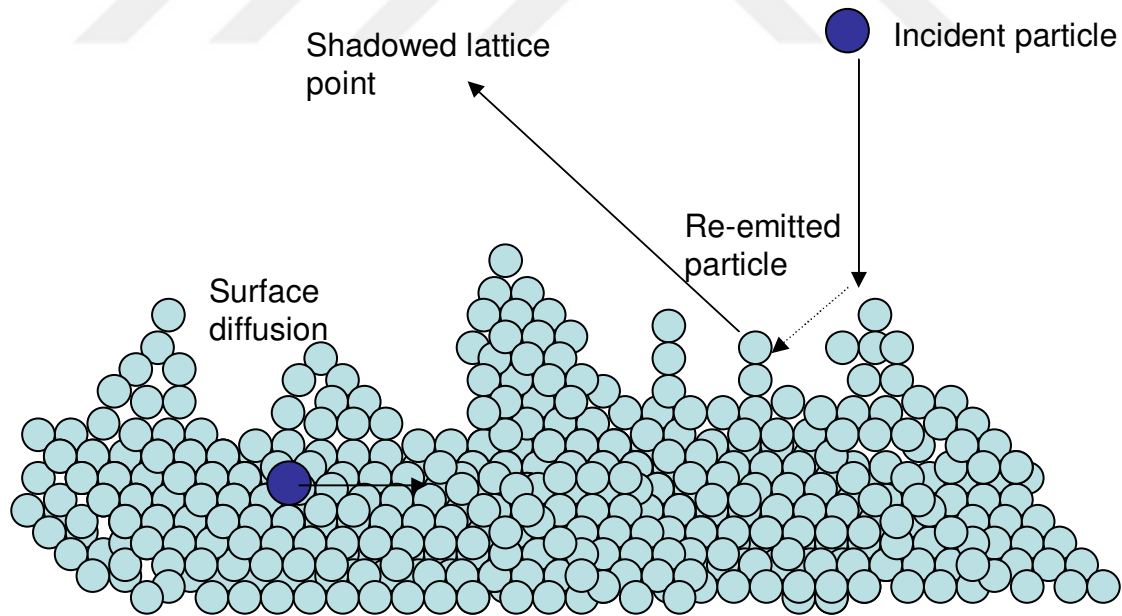


Figure 2.9: Illustration of Monte Carlo (MC) simulation method for thin film growth modeling is shown.

2.6. Thermal Evaporation Deposition

In this study, we have mentioned about physical vapor deposition (PVD) whose coating method involves purely physical processes such as high temperature vacuum evaporation or plasma sputtering bombardment rather than involving a chemical reaction at the surface to be coated as in CVD⁸⁵. We also have mentioned about some types of PVD and said that thermal evaporation is one of these types.

“One of the most common and simplest methods of thin film deposition is thermal evaporation deposition.”⁸⁵ To coat by thermal evaporation method, we often use thermal sources. “These sources could be hot filament, boat, open crucible sources, metal wire pieces, effusion cell, and powerful electron beam evaporator. Materials which are evaporated from hot filaments, boat and open crucible sources, are resistively heated. Metal wire pieces are hung from the filament, which melts and then evaporates them.”⁸⁵ “The electron beam gun is rapidly becoming the work horse of the thin film industry.”⁸⁶

Evaporation involves two basic processes: A hot source material evaporates and condenses on the substrate.⁸⁷ Therefore, thermal evaporation is often taken to be synonymous with resistive evaporation. When a high current is passed through a resistive element such as a light bulb, the filament gets white hot. Use a heavier filament and hook a few pieces of metal wire such as gold or aluminum and the metal wire will first melt and flow along the filament and then evaporate. This is the crudest but most common form of resistive evaporation.⁸⁵

In E-beam evaporation system, there are sources that heat the source material through bombardment with energetic electrons, which are emitted from a hot filament. An electron beam evaporator fires a high-energy beam from an electron gun to boil a

small spot of material. Since the heating is not uniform, lower vapor pressure materials can be deposited.⁸⁵ You can see 4 slots in E-beam evaporation system and we usually do not put substances alone. We put them in a crucible and we put those crucibles in those slots.

In Fig. 2.1, the source material is placed in a crucible and then heated until it evaporates and condenses on a substrate located above the crucible.⁵³ There are some advantages and disadvantages of thermal evaporation method compared to other deposition methods. For instance, in the overall process, using sputtering system takes more time than using evaporation. However, this depends on which source you use. One other advantage of thermal evaporation to sputtering deposition is⁸⁸⁻⁹⁶ that sputtering uses plasma, which produces many high speed atoms that bombard the substrate and may damage it. Evaporated atoms have a Maxwellian energy distribution, determined by the temperature of the source which reduces the number of high speed atoms. Sputter deposition and chemical vapor deposition have better step coverage but it may be advantageous or disadvantageous. It depends on the desired result.⁸⁵

3. LITERATURE SURVEY

There is vast amount of previous literature on thin film growth however in this chapter we will focus on the relevant ones to our work.

3.1 Literature Survey about Thin Film Deposition

3.1.1 Literature Survey about Sputter Deposition and Chemical Vapor Deposition

Drotar *et al.* examined the roughening behavior of the re-emission model for chemical vapor deposition. They have focused on pure first order re-emission and found that the interface roughness logarithmically with time and the scaling exponents are close to the Edwards-Wilkinson Model whose $\alpha=0$, $\beta=0$ and $z=2$. According to this model, $z \sim 2.33$. They then compared their results to experimental results on CVD.⁵⁵

This study is very important since we can learn about α , β , z for different models such as random deposition, Edwards-Wilkinson, KPZ, Mullins diffusion, shadowing growth and so on. Doing so, we can also learn whether it is defined. This paper also shows theoretical results for α , β , and z for thermal evaporation deposition for first order re-emission and we will compare our results with these values.

Drotar *et al.* examined the roughening behavior of a shadowing model. In that study, their results show that the roughening growth exponent $\beta=1$ for deposition and $\beta=0$ for etching. They also found that $z \sim 1.08$. This z is valid for deposition.⁵¹

Zhao *et al.* assumed a re-emission model and they have studied about the effect of re-emission model on the surface morphology. They have done their studies in low pressure chemical vapor deposition process.⁵⁶ In this paper, we can also see the importance of all order re-emission. First, they calculated their results in first order re-

emission and they got their results in all order re-emission and they compared the results in a graphic as shown below.

So far, we have surveyed different α , β and z values. However, people usually didn't measure their research by using re-emission. Xu and Goodman measured the initial sticking coefficient of copper on a thin silica film in the temperature range 90 and 700 K. They have observed that the copper sticking coefficient, S_c , depends on surface temperature and at low temperatures such as 90 K, 0.6 ± 0.1 . Above 400 K, S_c is less than 0.1. They also claim that upon covering the silica surface with multilayer water, the sticking probability increases to ~ 1 .⁷⁷

Other research is about sputtering system which was done by Karabacak *et al.* They investigated the growth front roughness of amorphous silicon films grown by dc magnetron sputtering at low pressure using atomic force microscopy. They have calculated $\beta = 0.41$, $1/z = 0.42$, and $\alpha = 0.83$. They have claimed that none of the known growth models can be used to explain the scaling exponents they obtained. They have extracted the scaling exponents and suggested a re-emission model to describe the possible surface growth mechanisms during sputter deposition.⁶⁷ Different than the re-emission model proposed Jason *et al.*,^{50, 77} the model they have used takes into account surface diffusion and has a more directional flux of particles compared to the one used in their work.

In their study, they have plotted β versus diffusion rate graphic. It is the number of diffusing particles per incident atom, D/F . Their graphic in their journal tells us, as D/F increases, β converges to the value ~ 0.34 . They have also measured that in the range of

high D/F , α approaches 0.38. They have fit the data by an exponential decay curve. In Fig. 3.1 and 3.2, we can see the range of experimentally found β values.

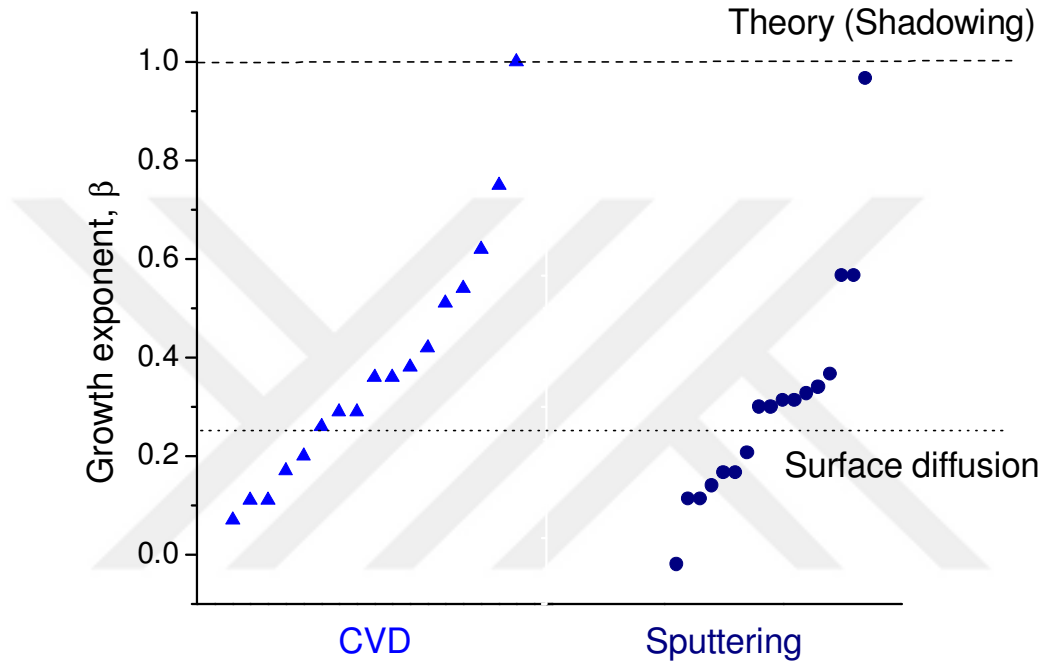


Figure 3.1: Experimental values of β reported in the literature for chemical vapor deposition and sputter deposition respectively.

In Fig. 3.1, people can see 16 different values for chemical vapor deposition, which are range from 0.13 to 1.0 and 17 different values for sputter deposition, which are ranging from 0.16 to 0.9. These values each indicate that there is a different research whose β 's are experimentally different. Some researchers found out why there is not a unique β . They have explained that the reason is re-emission effect.

3.1.2 Literature Survey about Thermal Evaporation Deposition

Thompson *et al.* thermally evaporated Ag/Si films at 300 K substrate temperature and found that $\alpha = 0.7$, $\beta = 0.26$, which is too close to Mullins diffusion model and $\xi \leq 100$ nm.^{64, 97} Palasantzas and Krim thermally evaporated Ag/quartz films at the same temperature and found that $\alpha = 0.82$, $\beta = 0.29$, and $z = 2.53$.⁶⁴

According to Chiarello's study, Au and quartz films are thermally evaporated at 500 K and they have found $\alpha = 0.95$ and $\xi = 330$ nm. They haven't observed a certain value for β .⁹⁸ He *et al.* evaporated Fe atoms at 333 K and found that $\alpha = 0.79$, $\beta = 0.22$.³⁹

Amaral *et al.* investigated the effect of evaporation on the interface roughness and mean interface height. They have done all their studies in (1+1) and (2+1) dimensions. For (1+1) dimensions, they have calculated $\alpha = 0.63$ and $1/z = 0.52$. Using the formula $\alpha / \beta = z$; we can assume that their β value is 0.33. As for (2+1) dimensions, their α and $1/z$ values are 0.43 and 0.31 respectively. Using the same formula, we can calculate $\beta = 0.14$.⁶⁵

Chiarello *et al.* has a study that is more focused on to find α value. They have carried out x-ray reflectivity and adsorption measurements on thermally evaporated silver and gold films deposited onto substrates held at 80, 300 and 500 K to investigate whether the surfaces of such films are fractal in nature. Their experiments are done 5° off normal incidence, because deposition at normal incidence produced no detectable increase in area. Their α results at 80, 300 and 500 K are 0.46, not fractal and 0.95 respectively.^{81, 98}

Yang *et al.* has reported a thin film growth morphology study of amorphous Si films using atomic force microscopy.⁵⁴ They have measured α and β , consistent with the surface diffusion model with noise. They have deposited Si on Si at $\sim -10^\circ$ substrate temperature.

Canas *et al.* has a study about relationships between the surface morphology and the height distribution curve in thermally evaporated gold films. Their β value is 0.38. They have done their experiments for different thicknesses.⁹⁹

Undoubtedly, these are not the all studies about surface morphology. According to Lutt's study, $\beta=0.1$, $1/z=0.6$ and $\alpha=0.23$. They compared their parameters with the KPZ equation.⁶² This is an important result for us since β is as low as 0.1. We can see that the growth exponent, β , is experimentally between 0.56 and 0.1. However, theoretical models say β is 0.25 with smoothening effect. So, why are the experimental results different? What model explains the reason for the difference? We are going to investigate those equations in this thesis.

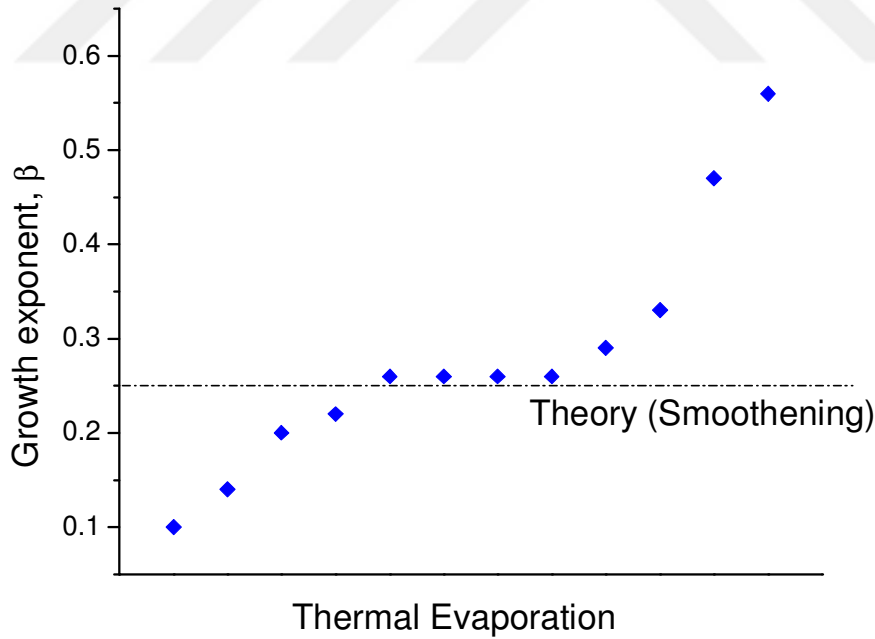


Figure 3.2: Experimental values of β reported in the literature for thermal evaporation deposition.

Fig. 3.2 depicts some growth exponent values for thermal evaporation deposition which are experimentally found by different researchers. Although theory says, β should be 0.25 with surface diffusion for thermal evaporation deposition; we have some values which are ranging from 0.1 to 0.56.

3.2 Literature Survey about Monte Carlo simulations

There are numerous studies which have been done using Monte Carlo (MC) simulations in every branch such as physics, chemistry, economy and biology. Here, we mentioned some of the studies which are related to my studies.

Drotar *et al*, examined roughening behavior of a re-emission model for chemical vapor deposition using MC simulations. They calculated their surfaces roughness parameters are close to the Edward-Wilkinson model. At last, they compared their MC simulation results to experimental results on chemical vapor deposition.⁷⁶

In 2000, Drotar *et al*, has another paper using MC simulations. They examined the roughening behavior of the shadowing model for 3D systems. Not only they worked deposition, but also etching. They also did some numerical studies and they compared their results with previous studies. They also compared their results with the experimental results which have been done by sputter deposition.⁵¹

Another important paper of Drotar *et al*, named ‘Mechanisms for plasma and reactive ion etch-front roughening’. The importance of their study is, using MC simulations; they calculated roughness exponent, growth exponent and the dynamic exponent which are universal. They mentioned universality towards the end of their study.⁵⁰

Karabacak *et al*, has some papers that data were obtained by using MC simulations. One of them is published in 2001 and named “Growth-front roughening in amorphous silicon films by sputtering”. They found $\beta=0.41$; $1/z=0.42$; and $\alpha=0.83$ in their studies.⁶⁷

In 2002, they have another published paper where they used MC simulations. This time their study is completed with plasma-enhanced chemical vapor deposition. Their α and β values are too close to their previous studies though their $1/z$ is somewhat different. Their approximate results are; $\beta\sim 0.40$, $\alpha\sim 0.77$, and $1/z\sim 0.28$.⁵⁵

3.3 Literature Survey about Numerical Solution Methods

There is not many studies about numerical solution methods as much as MC simulation method. There are some reasons of this situation. The most important one is numerical solutions are too slow and computers are not fast enough in the past. Even now, it is not fast enough.

One of the studies which is about numerical analysis but not the analysis of continuum equation, instead, Kuramoto-Sivashinsky equation is published by Drotar *et al*. This paper helps how to approach to a numerical analyzes. They calculated the initial-time values for β is around 0.22-0.25. They also calculated α , which is around 0.75-0.80, and the dynamic exponent, $z=3.0-4.0$.⁶⁰

The most related paper that is about the numerical solution of a continuum equation is Amar and Family’s paper. It is published in 1990 and the exact name of the paper is “Numerical solution of a continuum equation for interface growth in 2+1

dimensions”. It is also a rapid communications paper whose publishing process will be accelerated and gives extremely important results.⁷¹

In 1991, Toral *et al*, has a paper which is named, “Numerical study of a Langevin model for the growth of wetting layers”. They presented the results of a numerical study of a Langevin equation describing the growth of wetting layers. They focused on the layer thicknesses and density profile function parameters in their study.¹⁰⁰



4. GOALS AND OBJECTIVES OF THE THESIS

As presented in the introduction and review chapters above, the modeling of thin film growth process has been a challenging task due to the stochastic nature of the film deposition. As also presented in our literature review, it has been recently shown that only after inclusion of a re-emission mechanism of atoms during thin film deposition, growth models have successfully predicted the important surface roughness parameters of thin films produced by sputter deposition and chemical vapor deposition (CVD) techniques, which are among the most widely used physical vapor depositions techniques. The growth exponent β values (where growth exponent defines the dynamic evolution of RMS roughness with time according to a power law behavior $\alpha(t) \sim t^\beta$ and dynamic scaling theory requires $0 \leq \beta \leq 1$) reported for thermal evaporation deposition, which is another most well known thin film deposition technique, was in only partial agreement with the predictions of surface diffusion models, which state $\beta = 0.25$. Yet, a significant amount of thermally evaporated thin films were reported to produce β values ranging from 0.1 up to about 0.6. Although re-emission has been applied to sputter deposition and CVD growth modeling successfully predicting the dynamic morphological evolution, up to this date, no re-emission based modeling method has been developed for thermal evaporation growth. Therefore, we developed a new growth modeling approach for thermal evaporation thin film deposition process that will take into the effects of re-emission. The modeling method was based on Monte Carlo simulation techniques that includes the effects of re-emission, shadowing, surface diffusion, and noise. We also used numerical solution methods in order to reach our goals. In our study, simulated thin films were investigated for their dynamic evolution of surface morphology and roughness. In

addition, our work includes comparison of roughness evolution of experimentally grown and simulated thin films on patterned substrates in order to better reveal the possible effects or re-emission during thermal evaporation growth.

Below we present the objectives.

1. Development of Monte Carlo simulation codes for thin film evaporation deposition: MC codes developed for sputter deposition and CVD are already at hand, and these codes will be modified in order to simulate thermal evaporation process.

2. Investigating the effects of re-emission during thermal evaporation growth using Monte Carlo simulation methods: This is one of the biggest and most important parts of our study which includes detailed surface evolution analysis on simulated thin films.

3. Numerical Solution of a Continuum Growth Equation: In addition to evolution of surface morphology using MC simulation methods, we also analyzed the surface morphology by numerically solving a re-emission growth equation.

5. EFFECTS OF RE-EMISSION DURING THERMAL EVAPORATION GROWTH USING MONTE CARLO SIMULATION METHOD *WITHOUT* SURFACE DIFFUSION

5. 1 Background

As mentioned earlier, in order to simulate the thermal evaporation thin film growth, we used MC simulation methods. We used C++ code to do our simulations. There is a code available at hand previously developed for sputter deposition and CVD growth processes. This code was modified to include the growth parameters and mechanisms relevant to thermal evaporation deposition.

In MC simulations we used a 512*512 cubic lattice size substrate and all the particles are coming from top at normal incidence. We start from a flat surface and we can use different sticking coefficients, S_c , in our study. For our investigation, these coefficients vary from 0.1 to 1 whose increment is 0.1, and 0.9 to 1 whose increment is 0.01. First simulations, which are first order re-emission, do not include surface diffusion. We will do same simulations in all order re-emission where there is also no surface diffusion. We run each simulation 10 times with different random number generators and calculate their error bars. So, we give our outcomes in a range.

5.2 First Order Re-emission

As mentioned before, if the incident particle re-emits one time after hitting the surface is called first order re-emission. In Fig 5.1, we plotted root mean square (rms)

roughness vs. time graphic in the log-log scale. We used different sticking coefficients from 0.1 to 1.0 and whose increment is 0.1. The slope of the first stage of the graphic is 0.5 and this part is called random deposition. We can see that slope of the lines depend on their sticking coefficient. This slope is equal to the growth exponent. These growth exponents, β values, are given in Fig. 5.2. We can see that these β 's are between 0.07 and 0.5. For instance, $\beta=1$ for $S_c=1$ since there is no re-emission and it is randomly deposited. In Fig. 3.2, we mentioned about the experimental values of β reported in the literature for thermal evaporation deposition. We can predict the reason why people measured different β values reported in the literature is they may be used different sticking coefficients. Since we run our simulations using 10 different random number generators for each sticking coefficient, we plotted error bars in the same graphic in 5.2.

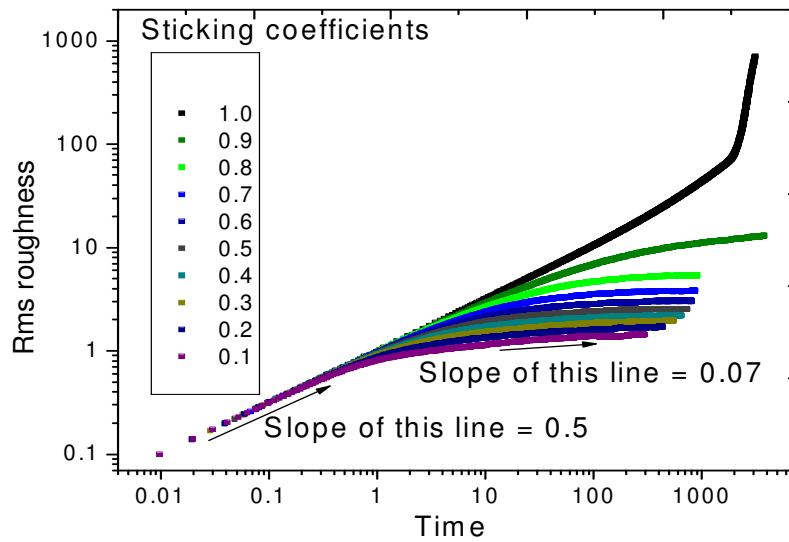


Figure 5.1: Rms roughness vs. time graphic when there is no surface diffusion in first order re-emission.

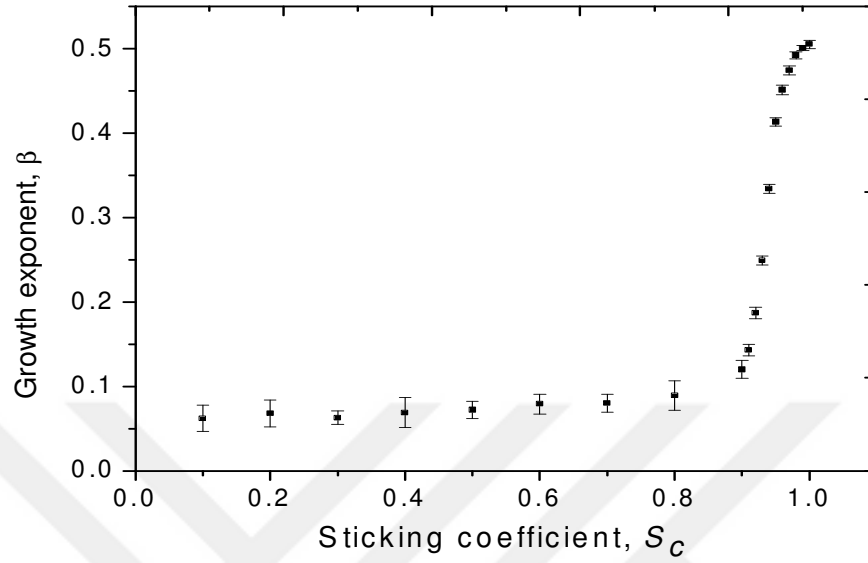


Figure 5.2: Growth exponent vs. sticking coefficient graphic when there is no surface diffusion in first order re-emission.

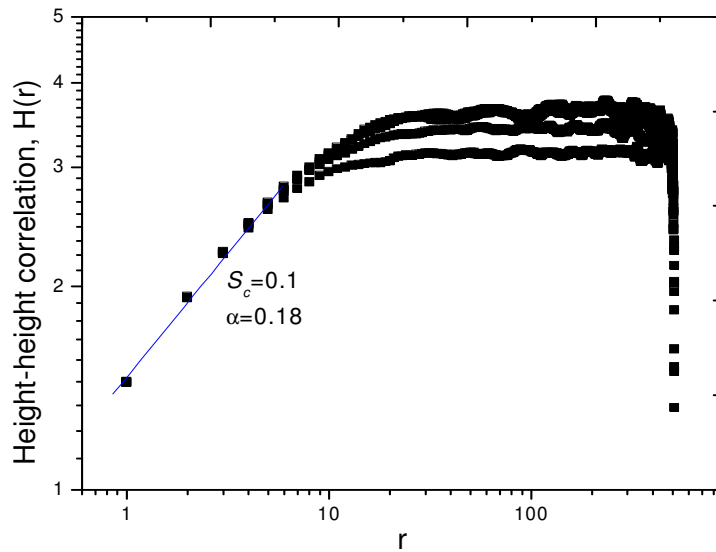


Figure 5.3: An example for how to find roughness exponent values. In this graphic the sticking coefficient that we arbitrarily used is 0.1.

In this graphic, the sticking coefficient is 0.1. The α value that we calculated is 0.18. To find this, we first plotted height-height correlation function vs. time graphics for some snapshots. These snapshots are not arbitrary. When we measure β values for a given sticking coefficient, we do not get fit for all the line. We choose this line where deposition occurs, which comes after the random deposition part and before the saturation part of the line. We calculate the times that correspond to linear fit line. Then, we plot height-height correlation function vs. r graphics at different times. Next, we calculate the slope of the average of these lines which is before the saturation part. This value gives 2α . In our simulations we usually calculated α values in a small scale due to the shortness of the time between random deposition and saturation.

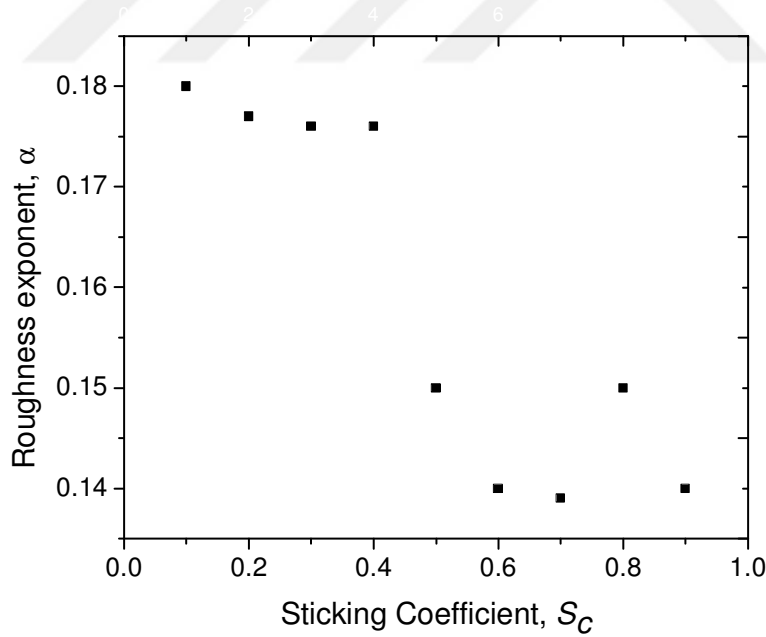


Figure 5.4: The values of roughness exponent for various sticking coefficients for first order re-emission and when there is no surface diffusion.

According to this graphic, we have two important results. First one is if the sticking coefficient is 1.0, in random deposition, we cannot define α . Therefore, it is not shown on the graph. The second important result is that although we can approximately predict sticking coefficients, S_c , just by looking β values, but we cannot predict S_c 's just by looking α values. As shown in Fig. 5.4, α values does not change in an order at different S_c 's.

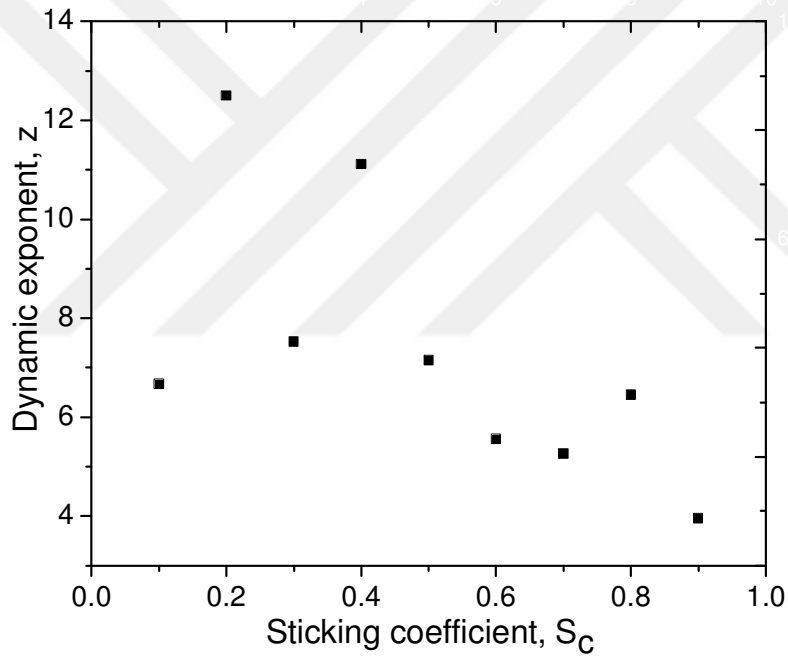


Figure 5.5: Dynamic exponent vs. sticking coefficient graphic of the first order re-emission when there is no surface diffusion for different sticking coefficients in the log-log scale.

As we mentioned in the second chapter, another important roughness parameter is the lateral correlation length (ξ). We can measure $1/z$ values from the slope of log-log plot of ξ versus t graphic. By taking the inverse of $1/z$, we can calculate z values.

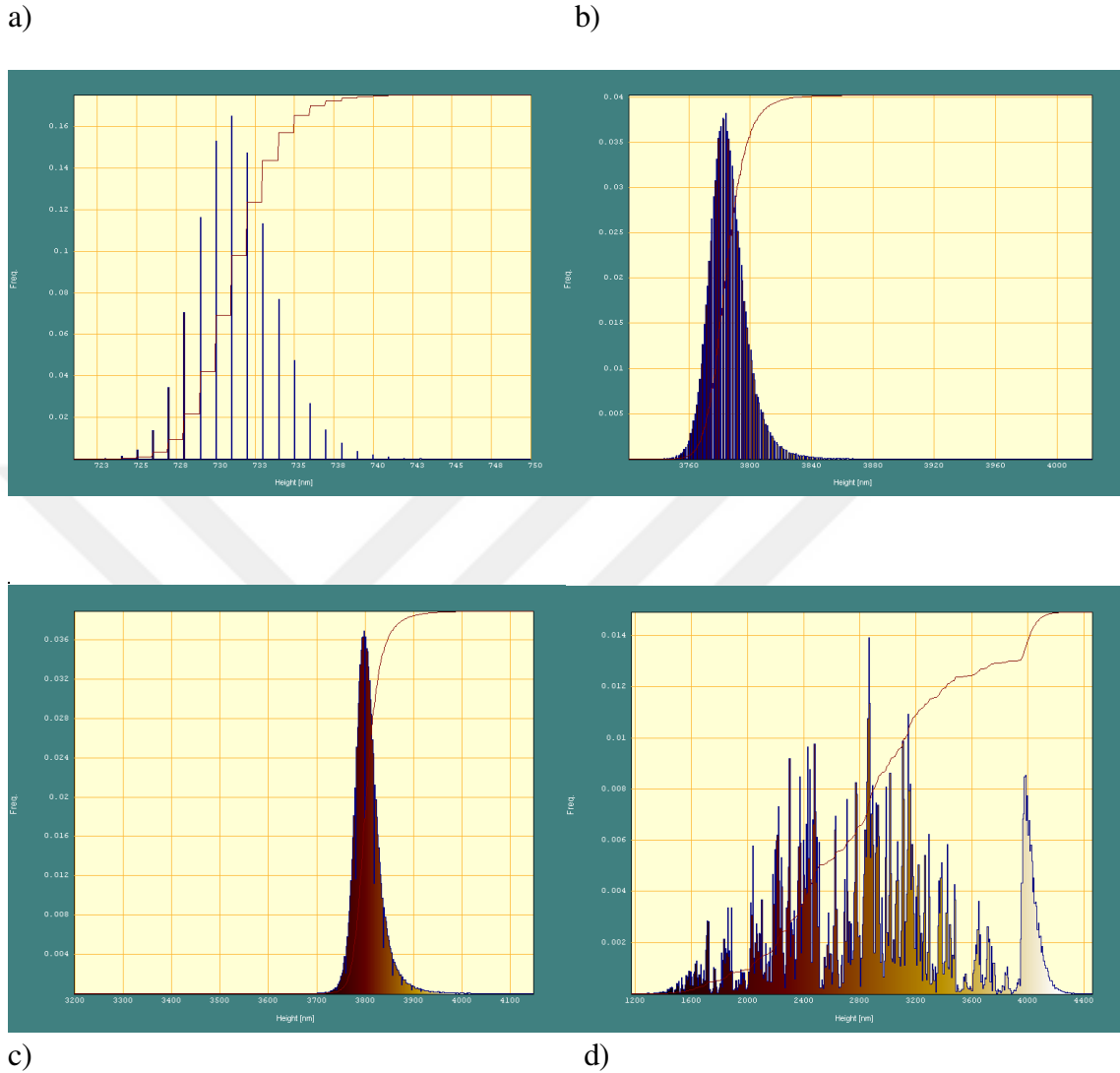


Figure 5.6 a- d: Height distributions of the first order re-emission graphic when there is no surface diffusion. These are the graphics for same amount of time whose sticking coefficients are 0.5, 0.9, 0.94, and 1.0 respectively.

When we examined height distribution data for all sticking coefficients, we see that at sticking coefficients which are lower than 0.95 show a Gaussian distribution. These are usually in a small height range. As for higher sticking coefficients, we see that there is a randomly frequented height distribution.

5.3 All Order Re-emission

As mentioned before, if the incident particle re-emits one time after hitting the surface, it is called first order re-emission. If this process happens multiple times, then it is called all order re-emission. When we examined Fig. 5.7 as shown, we realize that rms roughness vs. time graphics are almost same as Fig. 5.1 which are plotted for first order re-emission. These results affect β vs. S_c graphics; therefore, Fig. 5.8 gives almost the same results with Fig. 5.2. S_c values that are used for Fig. 5.7 and Fig. 5.8 are the same as Fig. 5.1 and Fig. 5.2. The reason why we used S_c 's which are from 0.9 to 1.0 is that there is a huge difference between these S_c 's. For other S_c 's, β values are close to each other, so it is predictable that the approximate value of β 's for any S_c which are lower than 0.9.

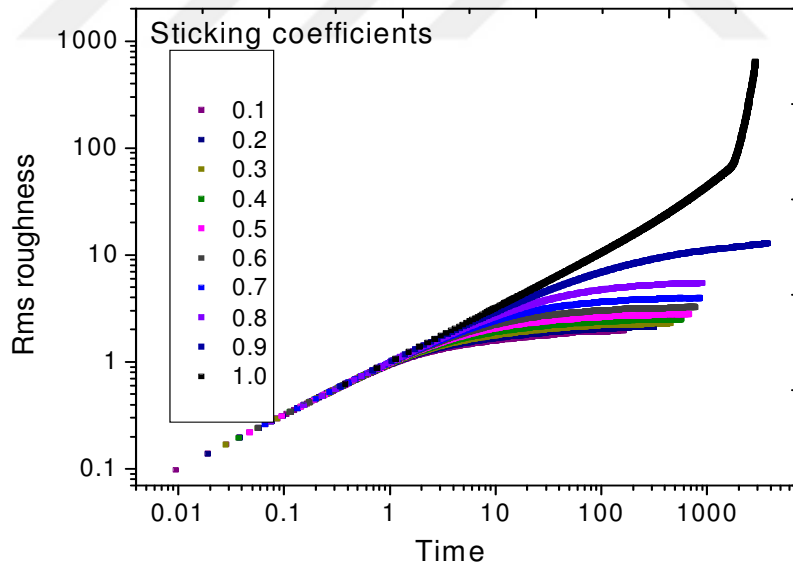


Figure 5.7: Rms roughness vs. time graphic when there is no surface diffusion in all order re-emission.

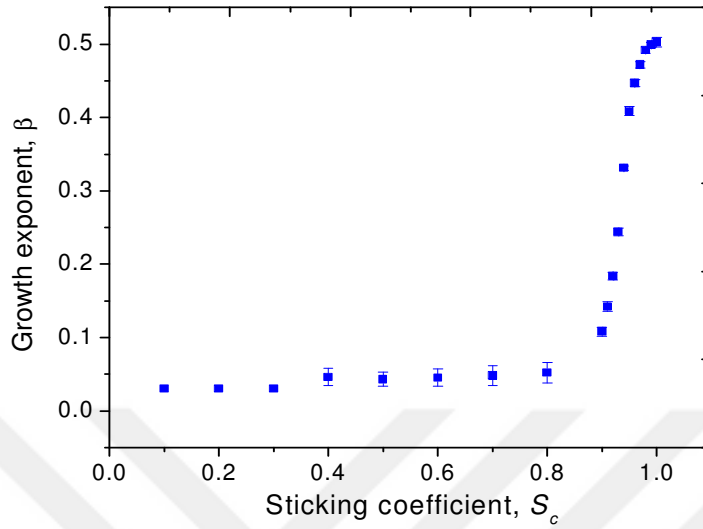


Figure 5.8: Growth exponent vs. sticking coefficient graphic when there is no surface diffusion in first order re-emission.

In Fig. 5.9 a- d, we see that the height distributions of all order re-emission when there is no surface diffusion. Results are similar to first order ones, at least they are Gaussian at S_c 's which are lower than 0.96. At S_c 's which are too close to 1.0, we see that there is a randomly frequented height distribution.

For higher S_c 's, we examined height distributions at different times with same sticking coefficients. We get similar results. Initially, there is Gaussian distribution and as time passes, we see deformation in the distribution.

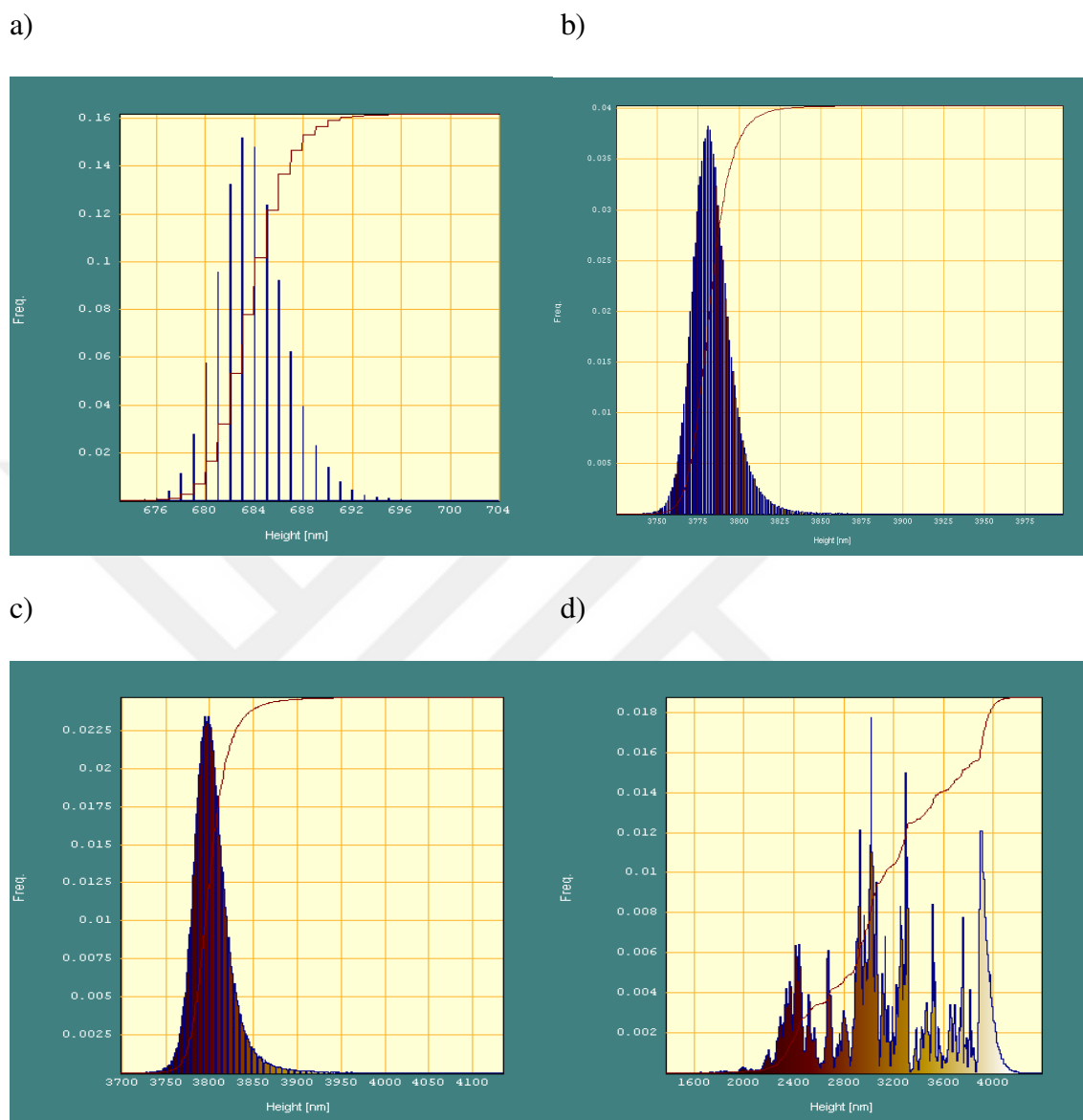


Figure 5.9 a- d: Height distributions of the all order re-emission graphic when there is no surface diffusion. These are the graphics for same amount of time whose sticking coefficients are 0.5, 0.9, 0.93, and 0.98 respectively.

5.4 Comparison between First Order and All Order Re-emission

Blue marks on Fig. 5.10 show β vs. S_c graphic for first order re-emission. Blue marks on Fig. 5.10 also show β vs. S_c graphic for all order re-emission which does not include surface diffusion. As you see in Fig.5.10, the graphic for first order and the graphic for all order are nearly the same and there is no significant change. At lower S_c 's, β 's for first order re-emissions are slightly larger than β 's for all order re-emission. However, this difference gets smaller at higher S_c 's.

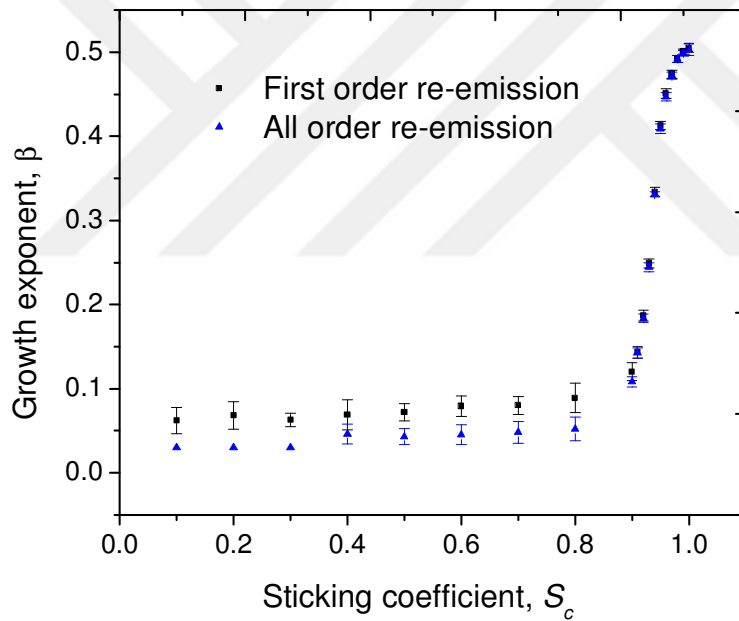


Figure 5.10: Comparison of growth exponent vs. sticking coefficient graphic for first order and all order re-emission in the same graphic which does not include surface diffusion.

The next two pictures show the comparison of the morphology whose S_c 's are same and 0.9 and they do not include surface diffusion, but one of them is for first order and latter is for all order re-emission.

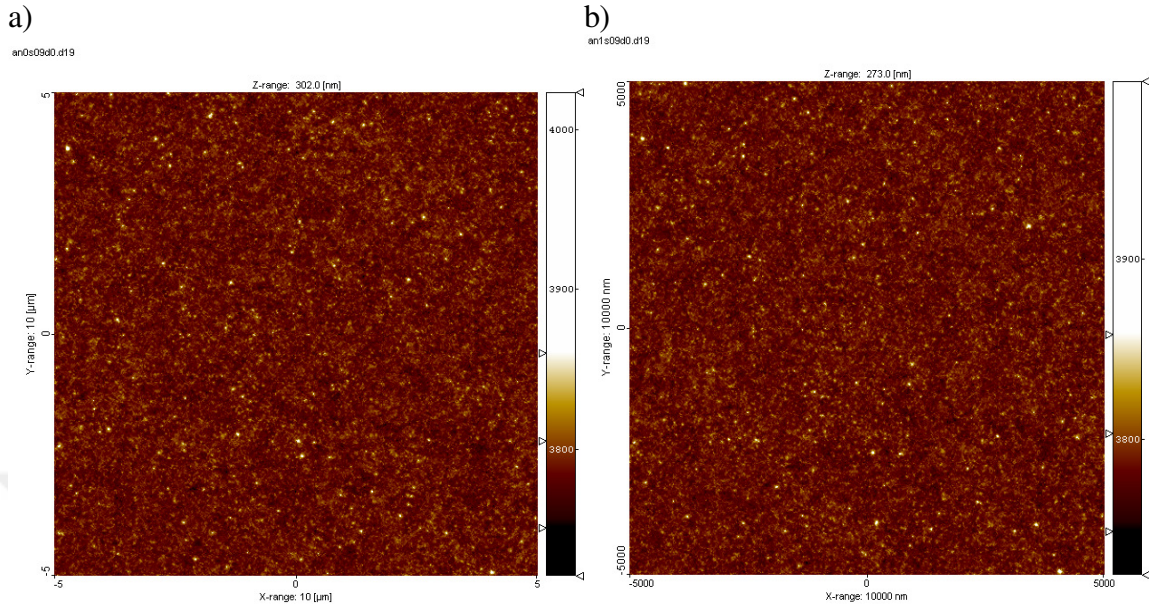


Figure 5.11.a, b: Image of the surface whose S_c is 0.9 and does not include surface diffusion in a) first order re-emission b) all order re-emission.

In Fig. 5.11, the vertical bar at the right side gives height information about that surface. Each height (nm) corresponds to a color. As height increases, color becomes whiter. On the right side of the bar, there are two arrows which denote the maximum and minimum heights at this surface. In the middle of the left side of the bar, there is an arrow at 3807 nm. This shows the average height. The other two arrows show that there is a frequency at those heights. As explained, there is no significant change between Fig. 5.11.a and 5.11.b.

Both types of re-emission have similar height distribution. They are not as close to their β values, however, they are both gaussian at lower S_c 's and their height scale for any given S_c is small.

5.5 Concluding Remarks

Our main goal was to find the effects of surface growth mechanisms. In order to reach our goals, we did some simulations which helps to reach our goals. In Fig. 5.2 and Fig. 5.8, we used S_c 's which are from 0.1 to 1.0 with 0.1 increments and from 0.9 to 1.0 with 0.01 increments. We repeated each simulation 10 times, so that people can see error bars on the graph. When the S_c is 1.0, our β value is 0.5. This matches with theoretical results which is random deposition. From 1.0 to 0.9, there is a sudden decrease which shows the effect of re-emission is very important. Between 0.9 and 0.1, β values do not change considerably since it is already very low. People may assume that 0.9 sticking coefficient is a high S_c , however, in our opinion, it is a low S_c due to β is decreasing from 0.5 to 0.1. Therefore, if the S_c is 0.9, it will show a very different surface morphology than from if the surface diffusion is 1 even if all other parameters are same.

We plotted β versus S_c graphic under the effects of first order and all order re-emission. Defining other morphological terms, such as α , z was our next step. We calculated how these terms differ with S_c on a graph under both the effects of all order and first order re-emission. We compared all our results with what theory predicts.

We also found out the height distributions of the surfaces after first and all order re-emission which was usually Gaussian if S_c is less than 0.96. PSD analysis and wavelength selection was done in first order re-emission and all order re-emission. Even though surface heights are not significantly correlated, they might exhibit a periodic behavior on a length scale larger than the lateral correlation length, but they did not.

Now, let us compare the surface morphology at lower and higher S_c 's. In Fig. 5.12 a, b, and c; we gave the surface morphologies of first order re-emission with surface

diffusion at higher and lower sticking coefficients. If we change the parameters from first order re-emission to all order re-emission, morphologies are approximately the same but not equal.

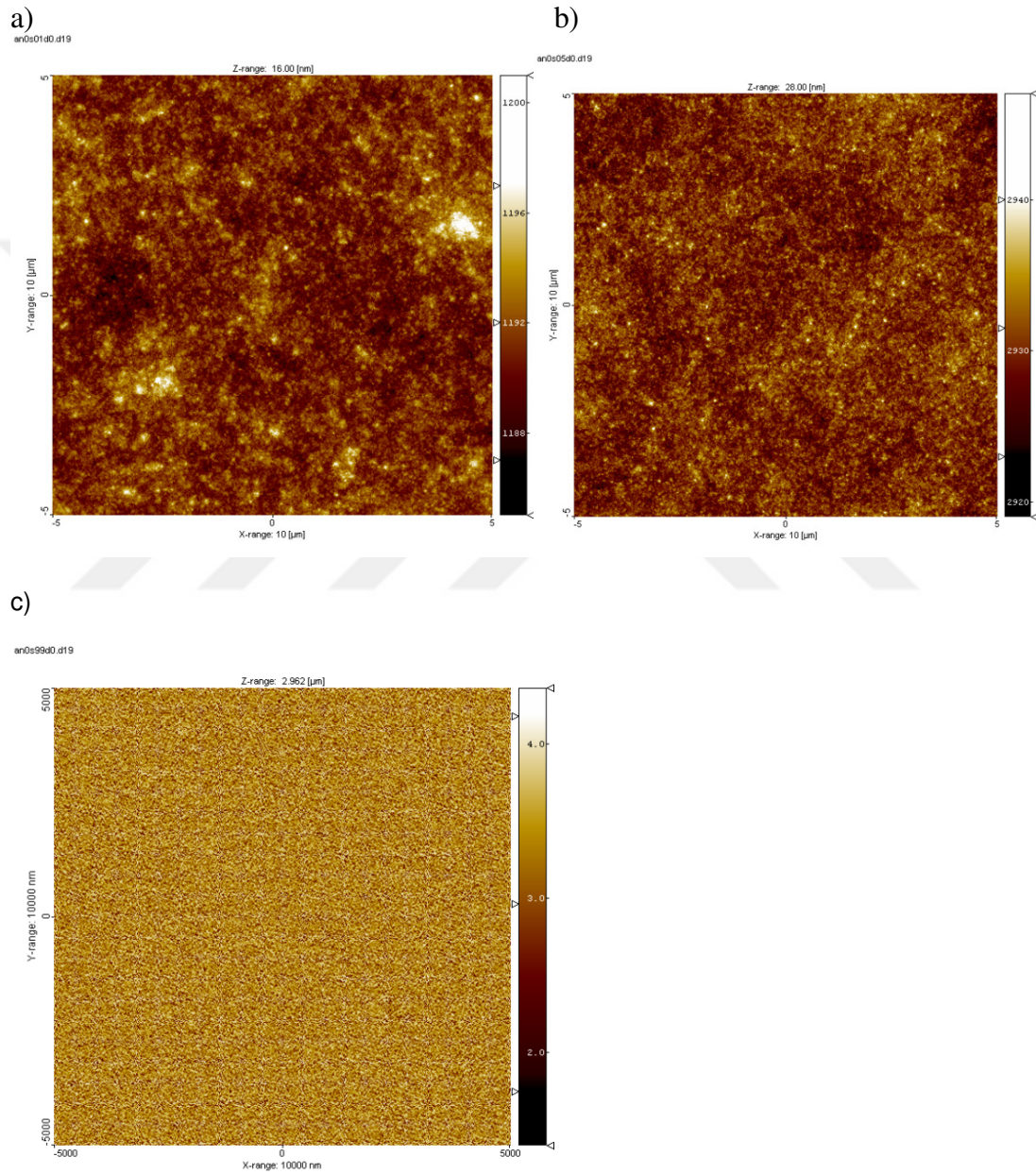


Figure 5.12 a-c: Image of the surface where sticking coefficients are 0.1, 0.5 and 0.99 respectively. These are all first order re-emission image where there is no surface diffusion.

In Fig. 5.12 a-c, we see that there is a huge difference if there is re-emission, even if all properties are same but re-emission exists. While in images 5.12.a and 5.12.b, the average heights are in nanometer scale, it is in micrometer scale in 5.12.c.



6. EFFECTS OF RE-EMISSION DURING THERMAL EVAPORATION GROWTH USING MONTE CARLO SIMULATION METHOD *WITH* SURFACE DIFFUSION

6.1 Background

As we mentioned before, if an atom comes and moves around nearby atoms before it sticks, we can say that there is surface diffusion. The diffusion rate, D/F , is also another important term, and the way to find the surface diffusion rate mathematically is explained below and in Appendix A.

Once particles are deposited, D/F atoms are randomly picked for a candidate to be diffused. This means not all of them are diffused. This is just the maximum number. D denotes the number of all surface atoms that are a candidate to be diffused. F denotes the number of deposited atoms after the surface diffusion process. The probability of an atom if it diffuses or not is $\exp(-E_b/kT)$ where $E_b = (n \cdot E_n) + E_0$. Here, E_0 is the force of the bond between the atom and under the atom, E_n is the force of bond between the atom and its neighbors such as under the atom, right and left side of the atom, k is Boltzmann constant, T is temperature, and E_b is the total binding energy.

When we include the surface diffusion in our simulations, we assume the diffusion rate is 350. The reason can be seen in Fig. 6.1. In this Fig., we see how growth exponent values, β , change as diffusion rate changes. As mentioned, the diffusion rate is the number of diffusing atoms per incident atoms. As discussed in the first chapter; according to Mullins diffusion equation, β will be 0.25 if the system includes the surface

diffusion. We already had results if the system has no surface diffusion, β is 0.5. We did the same simulations where the diffusion rate, D/F , is 50, 75, 100, 150, 200, 250, 300, 350, 400 and 450 respectively. We all used S_c is 1.0. The growth exponent values, β 's, in the graphic are decreasing as the diffusion rate is increasing. This is an anticipated result because the surface diffusion is a smoothing mechanism and small β denotes smooth surfaces. We get the diffusion rate where β is 0.25; that is what the theory says. The corresponding diffusion rate is 350. So, we have been doing all simulations which include surface diffusion when the diffusion rate is 350.

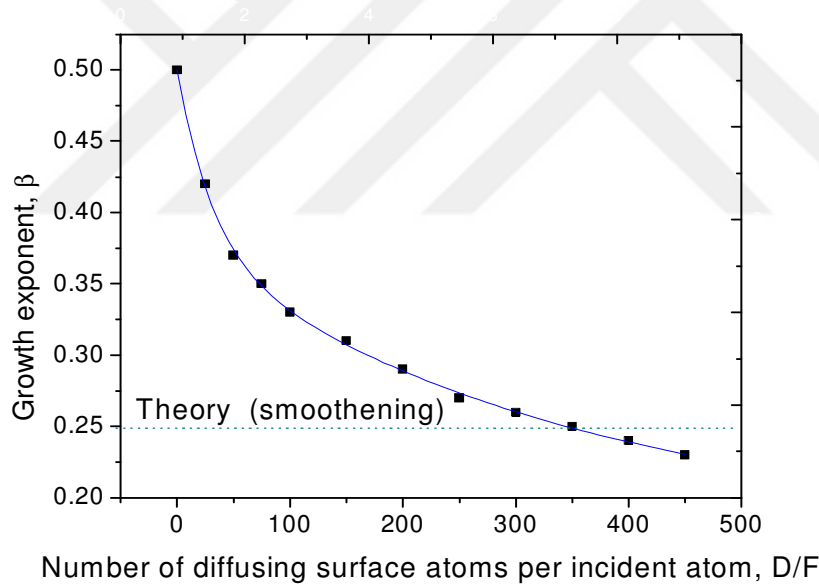


Figure 6.1: Growth exponent vs. diffusion rate graphic.

6.2 First Order Re-emission

So far we have run our simulations when there no surface diffusion. In this chapter, all our simulations include the surface diffusion process which has a smoothing mechanism. In Fig. 6.2, we plotted β vs. S_c graphic at first order re-emission

when surface diffusion exists. As we mentioned, the diffusion rate that we used is 350. If $S_c=1.0$; β is 0.25. This result matches with Mullins diffusion equation.

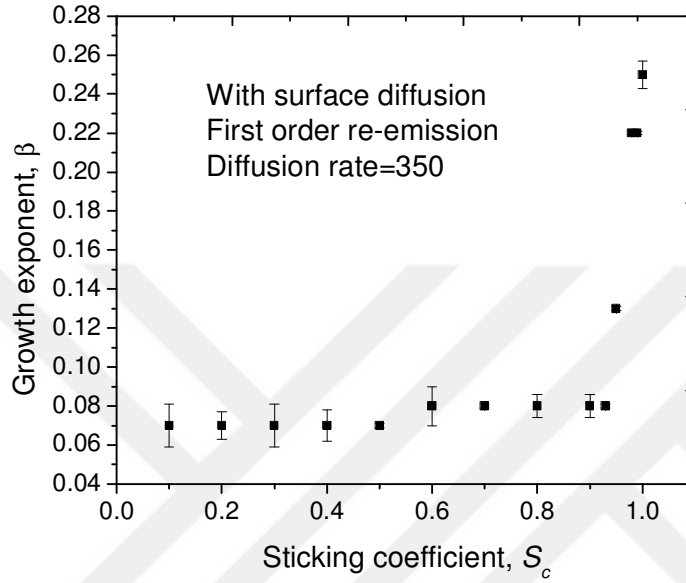


Figure 6.2: Growth exponent vs. sticking coefficient graphic at first order re-emission when there is surface diffusion.

According to this Fig., there is a sudden decrease between $S_c=1.0$ and 0.9 as well as for without surface diffusion case. There is also no significant change between $S_c=0.9$ and 0.1.

As for roughness exponent values, α , as in without surface diffusion case, there is not determined α values in an order. The only note that we can say is at $S_c=1.0$ we can define α as shown in Fig. 6.3. There is also not a predictable dynamic exponent, z , values just by looking S_c 's.

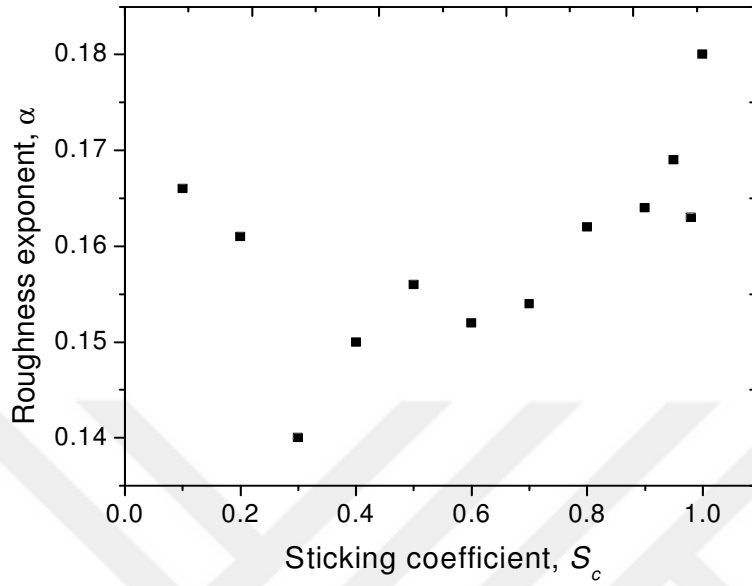


Figure 6.3: Roughness exponent vs. sticking coefficient graphic for first order re-emission when there is surface diffusion.

In Fig 6.4 a, b, and c, we see that there is completely a Gaussian height distribution for first order re-emission when there is surface diffusion. These graphics has all deposited with same amount of time.

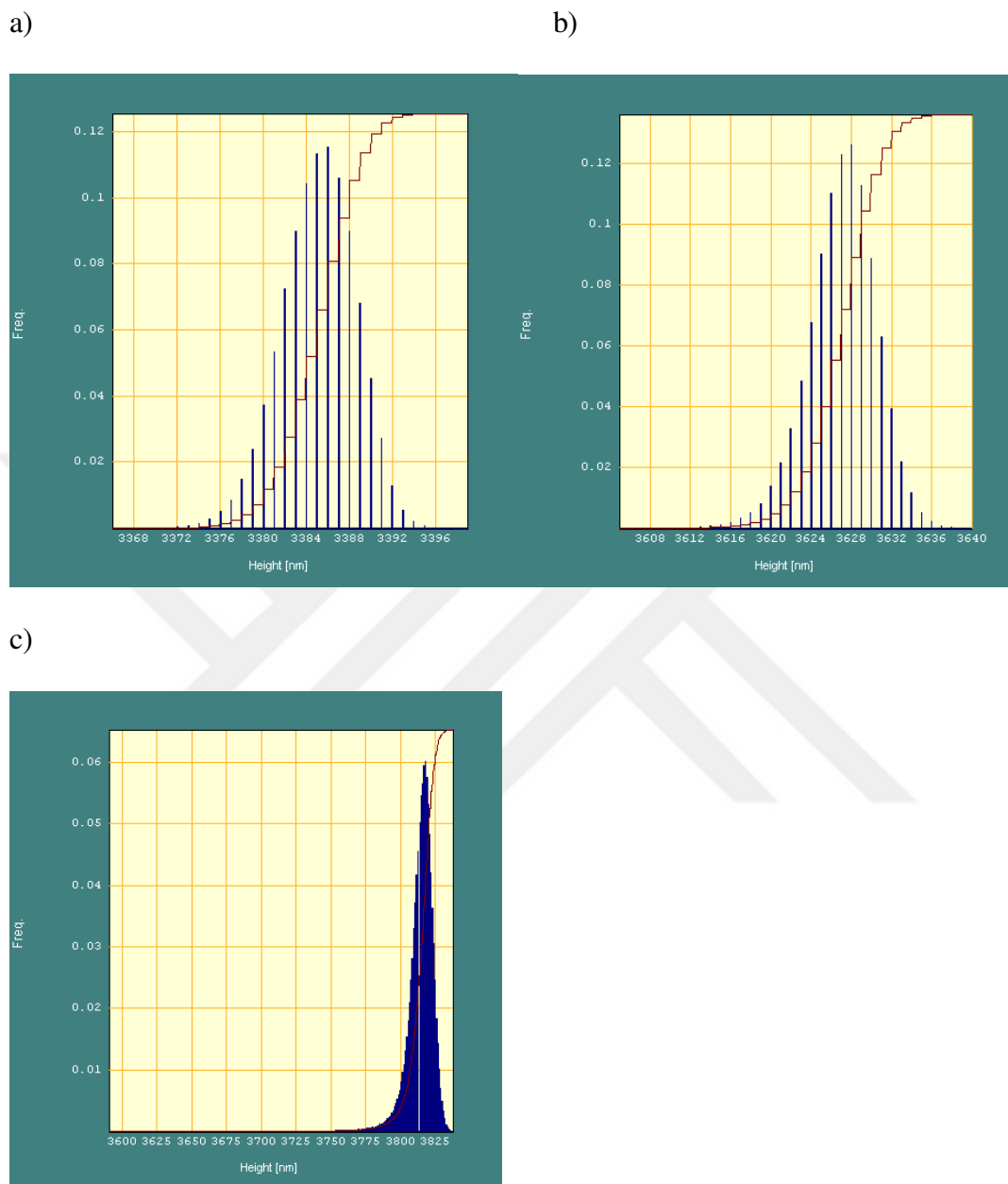


Figure 6.4 a-c: Height distributions of the first order re-emission graphic when there is surface diffusion. These are the graphics for same amount of time whose sticking coefficients are 0.8, 0.9, and 1.0 respectively.

6.3 All Order Re-emission

In Fig. 6.5, we plotted β vs. S_c graphic at all order re-emission when there is surface diffusion. We used the same diffusion rate, 350, for these Monte Carlo simulations. Like previous as in first order re-emission results, also like in all order re-emission but without surface diffusion case, we see that there is a sudden decrease between 0.9 and 1.0 sticking coefficients. Another point that we realized is, error bars are higher than previous results.

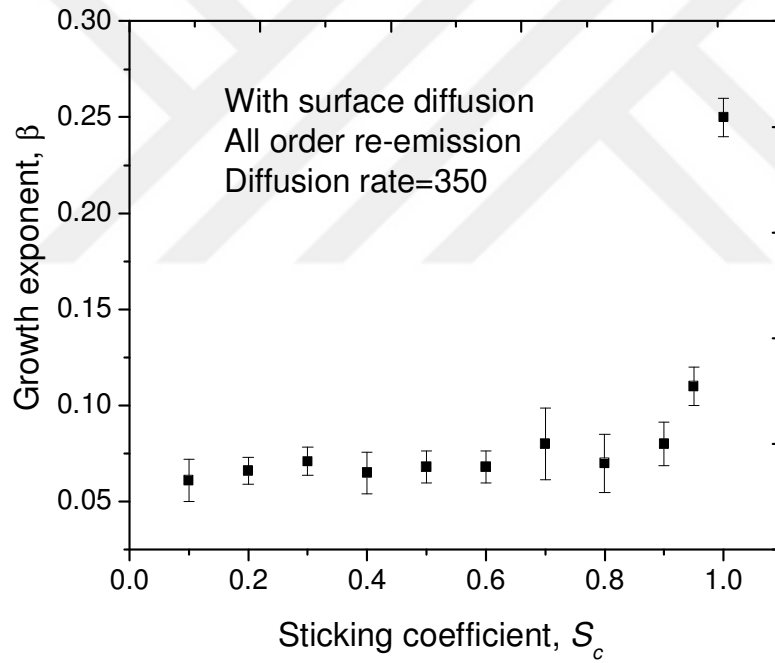


Figure 6.5: Growth exponent vs. sticking coefficient graphic at all order re-emission when there is surface diffusion.

6.4 Comparison between First Order and All Order Re-emission

We run the simulations for same parameters as in Fig. 5.10, but including surface diffusion and we investigated how β changes with surface diffusion in Fig. 6.6. We investigated that as in same figure; in Fig. 6.6, the β values are suddenly decreasing to a number, which is nearly 0.09 here and either slightly keeps on decreasing or remains same to 0.07. In Fig. 5.10, we also realized that there is no significant change for first order and all order re-emission for without surface diffusion case. This is valid for existing surface diffusion case.

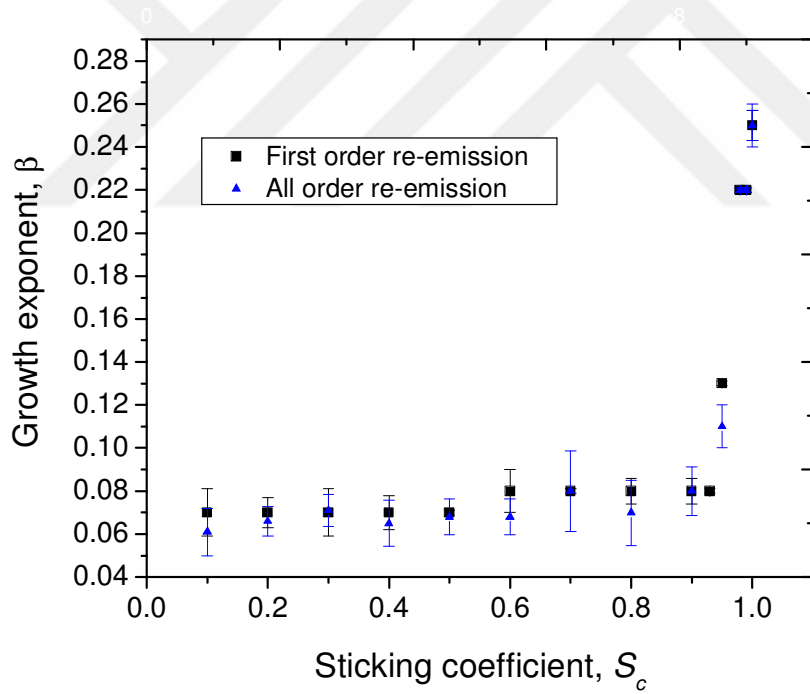


Figure 6.6: Comparison of β versus S_c graphic for first order and all order re-emission in the same graphic which involves surface diffusion.

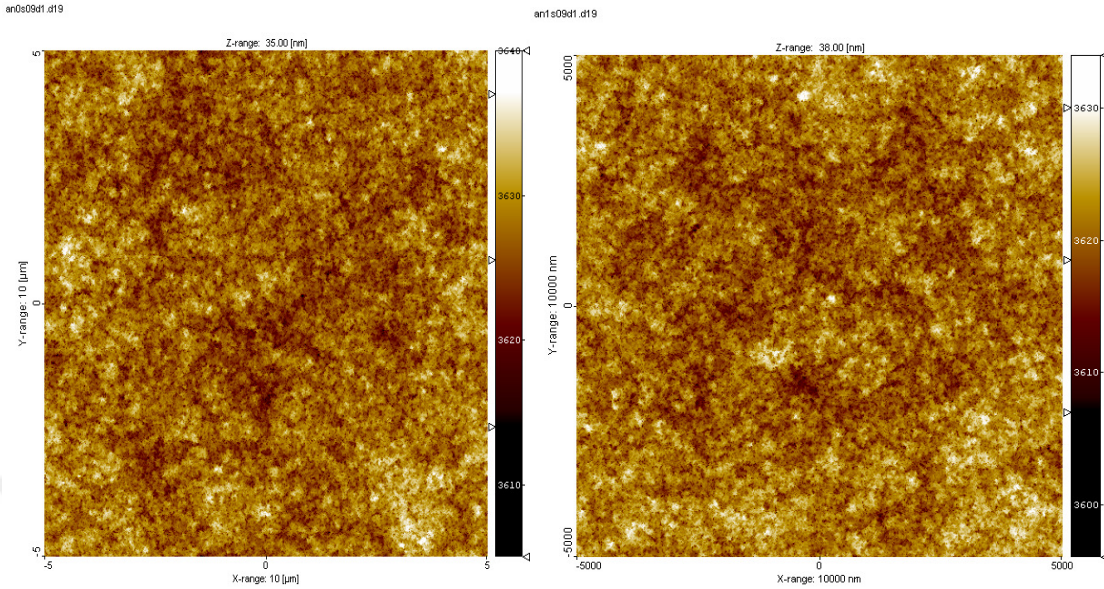


Figure 6.7 a, b: Image of the surface whose sticking coefficient is same, 0.9, and includes surface diffusion in a) first order re-emission b) all order re-emission.

Figure 6.7 is same with its S_c 's. As we said, there is no significant change based on the graphic, their morphology doesn't differ much. Different than Fig. 5.10, they are still first order and all order re-emission graphics but they include surface diffusion.

6.5 Competition between Re-emission and Surface Diffusion

Fig. 6.8 shows us β vs. S_c graphic for without surface diffusion and including surface diffusion cases. We just put the graphics for first order re-emission case since there is not a significant change between first order and all order re-emission. For without surface diffusion case, β is 0.5 when the S_c is 1. For surface diffusion case, β starts from 0.25 and keeps suddenly decreasing until the S_c is 0.9, and then keeps either slightly decreasing or remains same as in Fig. 5.2 and 6.2

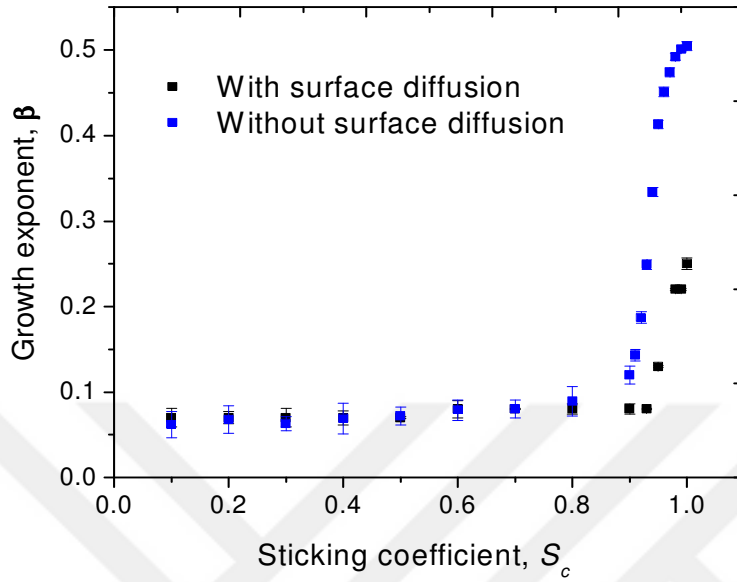


Figure 6.8: Comparison of first order re-emission vs. sticking coefficient graphics when involving surface diffusion and no surface diffusion case.

6.6 Concluding Remarks

In the first part of this chapter, we defined what surface diffusion and diffusion rate is. Then, we calculated which diffusion rate is best for our simulations. Next, we plotted for first order re-emission and all order re-emission graphics and compared themselves as was done in previous chapter. We briefly mentioned about their α values and height distributions graphics. At last, we compared surfaces of first and all order re-emission graphics at same S_c 's. Differences between these two types of re-emission are nearly same as in without surface diffusion case.

7: SURFACE EVOLUTION DURING GROWTH ON PATTERNED SUBSTRATES

7.1 Background

So far we have analyzed the surface morphology of flat substrates. We now calculated the surface morphology of patterned substrates. We used nanospheres as a substrate. The purpose of using nanospheres is to find out how the system changes if we use a rough surface. We are going to investigate the system if there is re-emission, if there is surface diffusion but not re-emission, and if there are both of re-emission and surface diffusion. Doing so, we can see the effects of re-emission and surface diffusion and we can decide which one is dominant. At the surface morphology whose substrate is smooth, we observed that re-emission has a dominant effect. So, we will see how the morphology changes if the substrate is patterned. All patterned surfaces are rough but not all rough surfaces are patterned.

Fig. 7.1 represents an example of initial patterned substrates. We used different radius. The reason why we used different radius will be explained in the following pages.

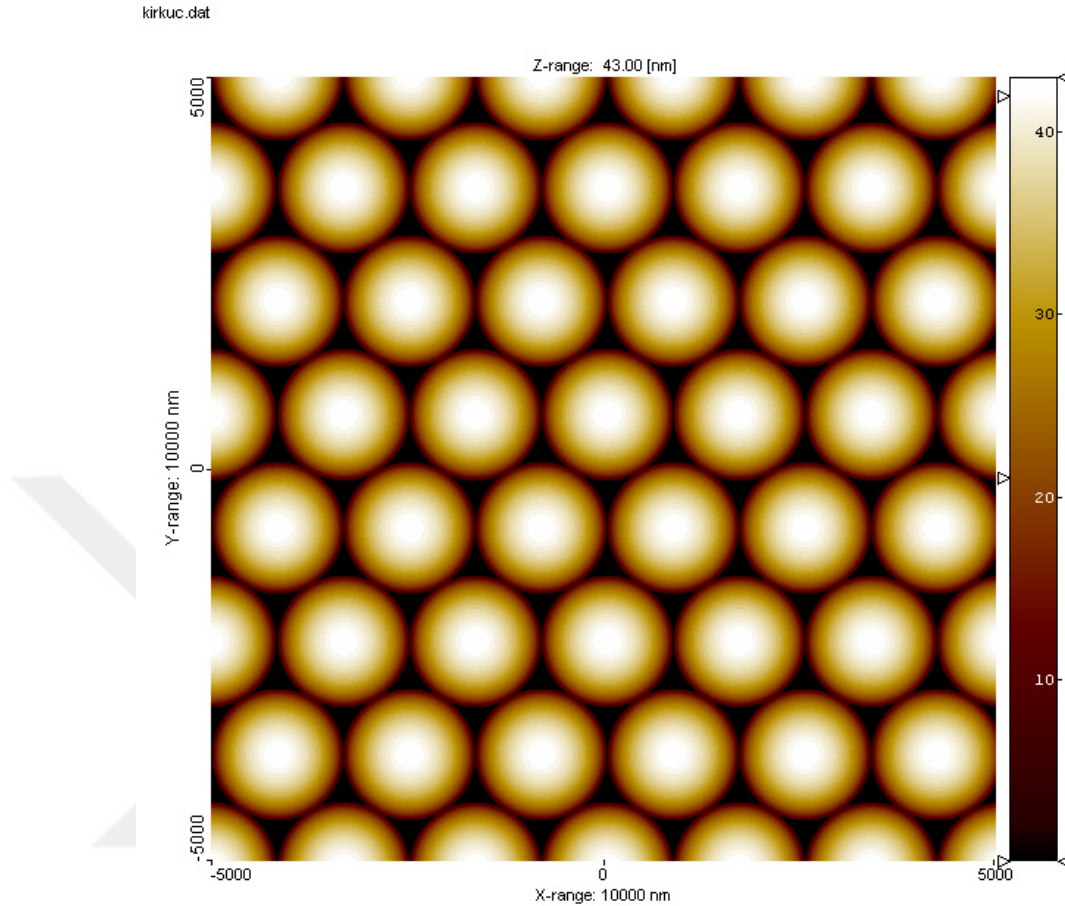


Figure 7.1: Top view of a patterned substrate. The circles represent nanospheres and they are in hemispherical-hexagonal form. The radius of these nanospheres is 43. The height of each point can be predictable by looking the scale at the right of the picture.

7.2 Surface Evolution with Patterned Substrate (For all Cases like Re-emission and Surface Diffusion)

Fig. 7.2 shows us rms roughness vs. time graphic whose simulations done in Monte Carlo simulation method for patterned surface where first order re-emission is used and not including surface diffusion.

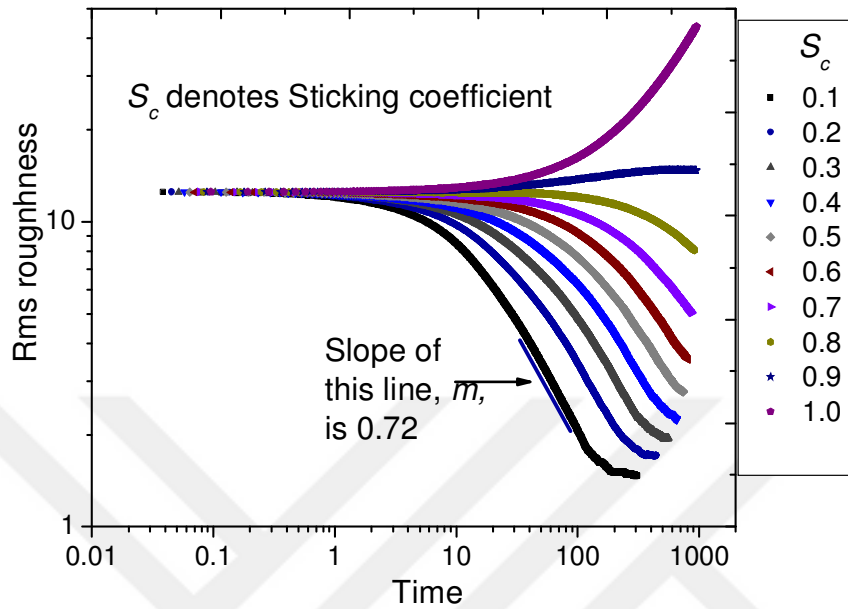


Figure 7.2: Root mean square roughness vs. time graphic in patterned surface. Each line corresponds sticking coefficients from 0.1 to 1.0, from bottom to top. No surface diffusion is used in this graphic. This graphic is for first order re-emission when there is no surface diffusion.

In Fig. 7.2, each line corresponds to one sticking coefficient which starts from 0.1 to 1.0 from and 0.1 incrementally from bottom to top. This is very important graphic that shows the effect of re-emission. So, if there is no re-emission there is no smoothening. There is a growth exponent which is 0.6 theoretically found by using Monte Carlo simulations. It can be seen that, as re-emission increases, smoothening increases. In Fig. 7.2, the most smoothening effect can be seen when S_c is 0.1.

In Fig. 7.3 a, b, c, and d, we see that how shapes of surfaces change as time passes. These images are obtained with STM calculator program. As you see in the picture, initially we have nanospheres and they are in hemispherical-hexagonal form. The

radius of these nanospheres is 43. Black color represents low-height areas; white color represents higher areas. After a while, incident atoms fill the lower surfaces since they come from the top in our simulations. As time passes, height difference between lower and higher areas decreases.

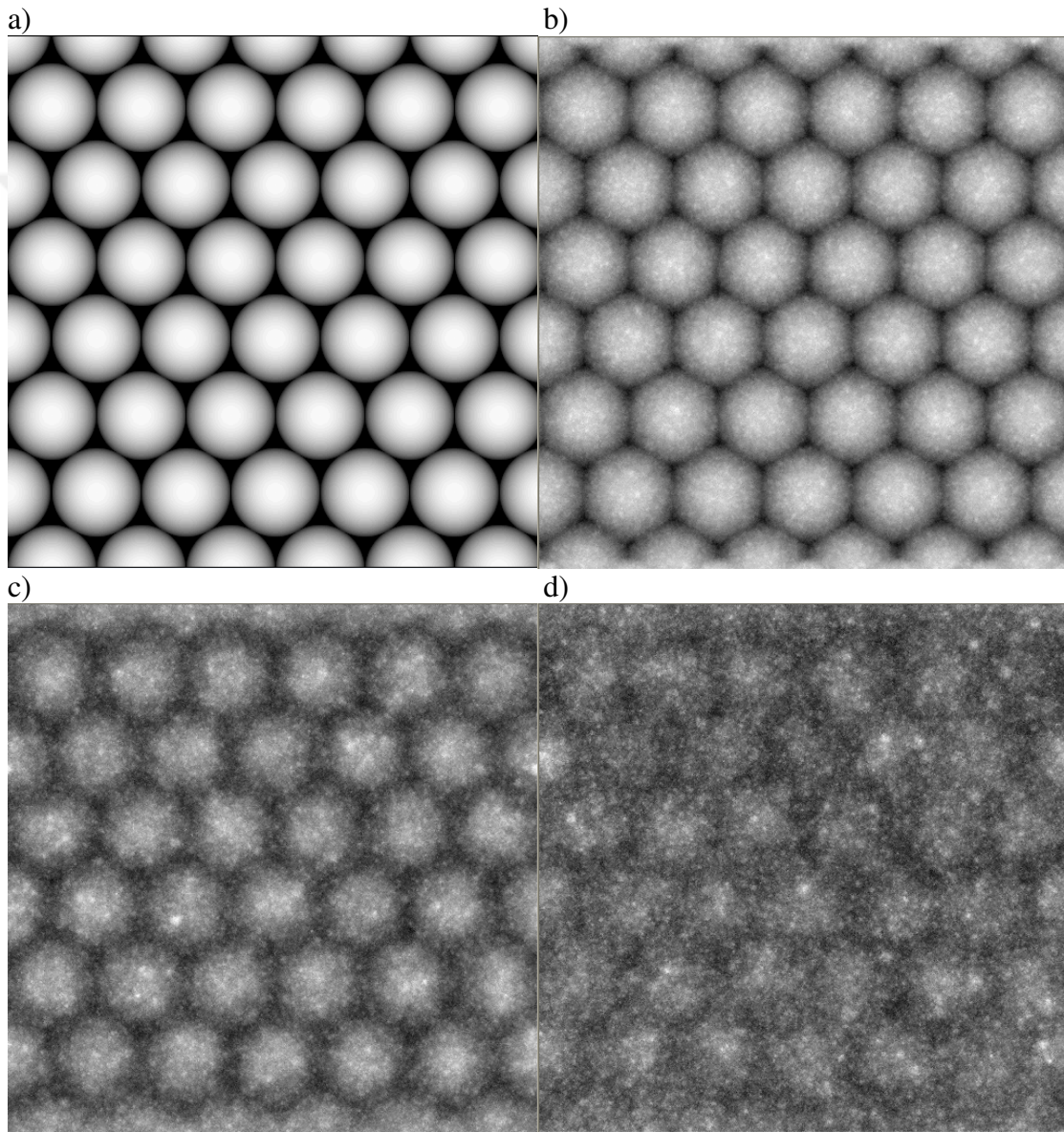


Figure 7.3 a-d: Images of the patterned surfaces whose all parameters are same but they are all in different times. This figure shows how substrates reshaped as time passes if we use patterned substrates.

In previous chapters, we explained what growth exponent, β , is and we explained how to calculate it. In this chapter, instead of growth exponent; we define *smoothing exponent*, m . It is same with growth exponent but it is the opposite of growth exponent. Although the slope is positive in growth exponent, it is negative in smoothing exponent, m . In Fig. 7.2 it can be seen how to calculate it. We used different radiuses since we want to see the effect of radius over morphology and if the smoothing exponents are same when the S_c 's are same, we can predict the S_c just by calculating m . Our results of different radiuses such as 16, 30, 43, and 60 are as shown in Fig. 7.4. One of the properties of our system is, it is 512*512 unit lattice size.

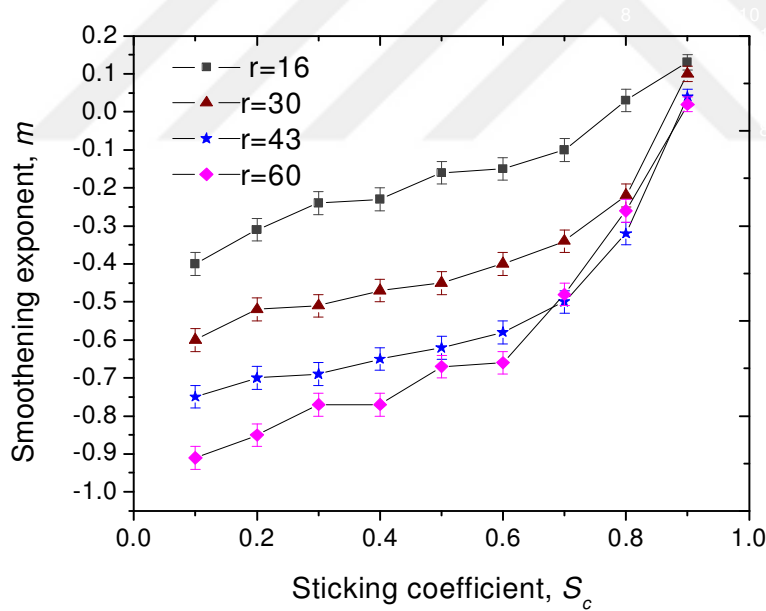


Figure 7.4: Smoothing exponent versus sticking coefficient graphic of a patterned surface at different graphics for different radiuses.

We see that m 's are different at different radiuses. If it would be same, we could have predicted S_c just measuring m 's. The way how we do is we are going to measure

some of these values experimentally and we can predict sticking coefficients by measuring m . We investigated a relationship between some certain radiuses such as 30 and 60; however, we could not find.

When we are calculating smoothening exponents, we first found rms roughness vs. time graphs. First, we subtracted mean heights of nanospheres as a bare substrate. For instance, mean height of the nanospheres, whose radius are 43.0000 units, are 25.6017 units. Then we plotted the log-log plot of rms vs. time graphs. Next we calculated the slope of smoothening part.

These graphics are for first order re-emission. We also have done at all order re-emission for radius 43 and we have seen that there is no significant effect for first order and all order re-emission. Remember that there has been a small effect when the substrate is flat. You can see this similarity for patterned surface in Fig. 7.5.

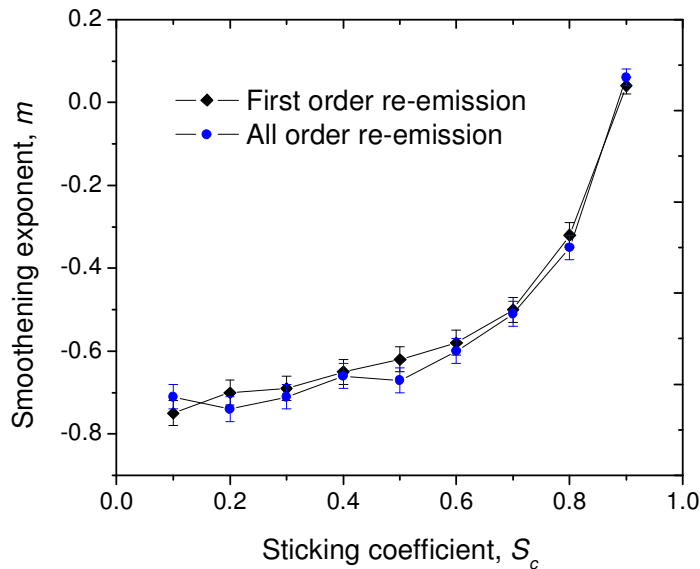


Figure 7.5: Comparison of first order and all order re-emission for patterned surface. This comparison is selected from where $r=43$.

We simulated all of the same simulations using all order re-emission while sticking coefficients are same, which starts from 0.1 to 1.0 and 0.1 incrementally. We plotted for first order re-emission and all order re-emission in the same graphic in Fig. 7.5. In this figure, black dots show the results for first order re-emission and blue dots show for all order re-emission. As you see, there is not much change. Although it seems a small difference, when we consider the noise effect, it is almost same.

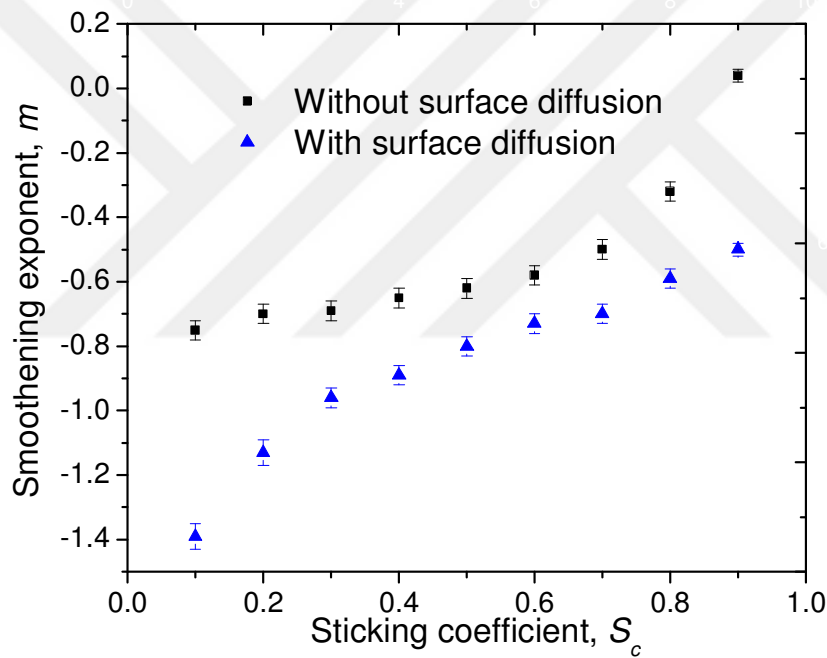


Figure 7.6: Comparison of with and without surface diffusion for $r=43$ at first order re-emission. This comparison is selected from where $r=43$.

Fig. 7.6 gives us the smoothing exponent versus sticking coefficient graphic of re-emission based model and surface diffusion based model. It is not shown on graphic but if $S_c=1.0$; there is no smoothing but growth exponents are approximately 0.03 in both cases.

We have not compared the same graphic for all order re-emission because when the initial surface is flat, we also have seen that there is not much change for patterned surface. You can see this in Fig. 7.5. Another important point here that should be taken into consideration so far is before calculating smoothening exponents, we calculated root mean square roughness by subtracting the mean height of the bare substrate.

When the initial surface is flat, remember that there is not much difference between first order and all order re-emission. There is some difference if S_c is more than 0.9 but it was almost same in lower S_c 's. If there is surface diffusion but not re-emission; we found out that surface diffusion has an effect but not as much as re-emission. As for patterned substrate case; there are some differences between the re-emission without surface diffusion case and re-emission with surface diffusion case. Even if surface diffusion has effect, it is not as dominant as re-emission as calculated. This dominant effect of re-emission is also expected in our experimental results. We also can say that if there is smoothening, re-emission exists. If there is no smoothening, re-emission is not important.

When substrate is flat and S_c is lower than 0.8, β 's are almost same. β does not depend on S_c . However, if substrate is patterned and S_c is lower than 0.8, m 's are not same. This does not matter for other parameters such as if there is surface diffusion or not; or if it is all order re-emission or first order re-emission.

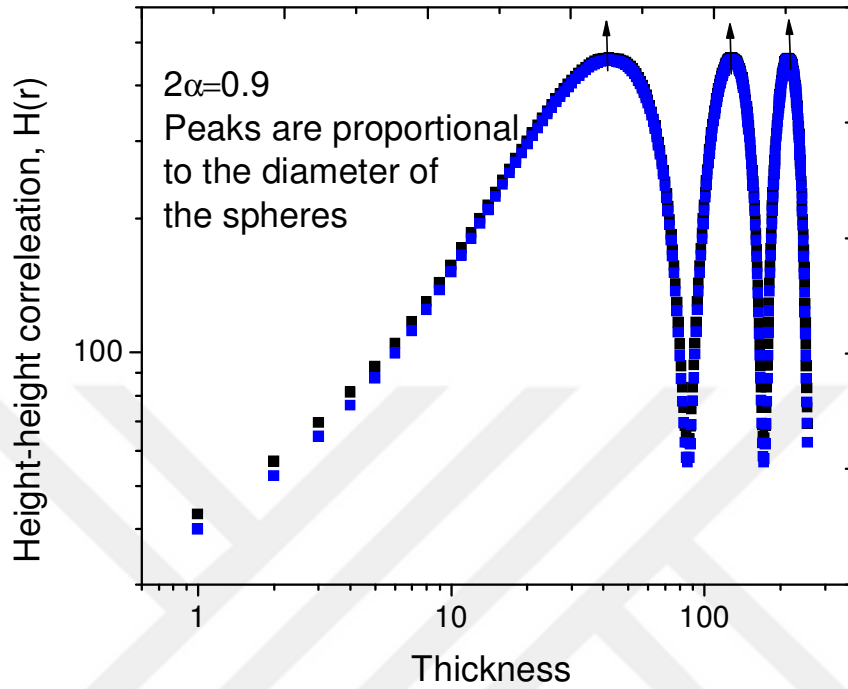


Figure 7.7: Height-height correlation function of a patterned surface whose radius of the nanospheres at the substrate is 43 and which has a surface diffusion but there is no re-emission.

Fig. 7.7 shows the height-height correlation versus thickness graph. The thickness of the peak points is proportional to the radius of sphere. Since the radius is 43 here, peak points are 43, $43*3$, $43*5$... So, we can say distances from each peak are 86. In this Fig., $2\alpha = 0.9$ and distance between peak points are proportional to the diameter of the nanospheres.

In this Fig., there are black and blue dots. These dots show this function at different times. At initial times, this graph holds. However, as time passes, this graph deteriorates. This deterioration has an effect on finding α and $1/z$. It makes impossible to find them.

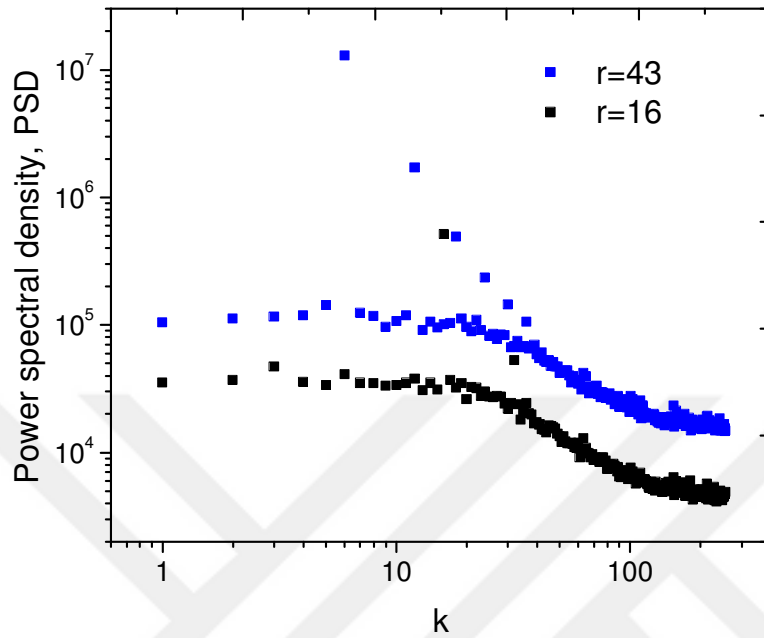


Figure 7.8: Power spectral density function of a patterned surface whose radius of the nanospheres at the substrate is 43 and 16, which has a surface diffusion but there is no re-emission.

In Fig. 7.8, we see that some peak points whose values are 6, 12, 18, namely factors of 6. Fig. 7.8 is different version of illustration for presenting power spectral density function. Peak points of k are the factor of 6. The number 6 can be calculated from $(\text{lattice size}/2*r) \sim 512 / (2*43)$. As mentioned in chapter 2.2, we can use wavelength selection by using PSD. The easy way to calculate λ is, $\lambda = 2*\pi / k_{\text{max}}$ where k_{max} the maximum peak frequency position.

7.3 Concluding Remarks

After finding the effects of re-emission and surface diffusion, and defining the morphological terms, we repeated all our simulations when substrate is patterned. This surface is a mounded surface where we can do wavelength selection easily. Normally, both re-emission and surface diffusion has a smoothening effect. We did all the simulations separately with a patterned substrate. In first case, we only included re-emission, and in the second case we included surface diffusion. In all our patterned substrates, we had nanospheres in 512*512 lattice size. First we used 43 diameter nanospheres; however, we did our simulations with different diameters and observed the difference. These differences are given in Fig. 7.4.

In order to determine this long range behavior the Power Spectral Density (PSD) function is used.³⁷ After finding PSD, which is the Fourier transform of the surface heights,⁵⁹ we made wavelength selection analysis.

8. A NUMERICAL SOLUTION APPROACH TO CONTINUUM GROWTH EQUATION

8.1. Background

In previous chapters, we have utilized Monte Carlo simulation methods in order to investigate dynamic evolution of surface roughness during thermal evaporation growth with re-emission effects included. Now, in this chapter, we will use an alternative approach for studying the same problem this time by numerical solution of a continuum growth equation. We will first provide a brief background on what numerical solution method is. Then, we are going to give the motivation for using numerical solution methods in this work. Next, we are going to present the -re-emission continuum growth equation- that we will try to solve. Last, we are going to mention about how we can solve continuum equation numerically and we will present our results.

“Numerical solution methods are a class of methods for solving a wide variety of mathematical problems. These problems can have their origins as mathematical models of physical situations. The combination of numerical solution methods and digital computers has created a tool of immense power in mathematical analysis. Numerical solution methods are capable of handling the nonlinearities, complex geometries, and large systems of coupled equations which are necessary for the accurate simulation of many real physical situations.”¹⁰¹ In the past, numerical solutions are not very common for hard problems since computers are slow. Since numerical solution methods are very easy and cheap and computers are fast, these methods are preferable.

Working with numerical solution methods is a good way to solve a problem.

There are some types of methods and people prefer to select the most accurate. Almost none of the methods about numerical solution methods give the exact result but some methods give most near results than others. There are also some effects that affect the accuracy of the result such as how big is our steps and how much iteration has there.

The formula that we are going to solve numerically is continuum equation

$$\frac{\partial h}{\partial t} = \nu \nabla^2 h - \kappa \nabla^4 h + \sqrt{1 + |\nabla h|^2} \left[\sum_{i=0}^{\infty} s_i F_i(\mathbf{r}, t) \right] + \eta.$$

ν is surface tension and κ is the surface diffusion term. η represents noise term. s for sticking coefficient and F for flux. Since we are going to solve a type of numerical solution of ordinary differential equations, we will first explain how to solve an ordinary differential equation numerically. There are some methods but the most common is the Euler method.

According to the idea of Euler method, how we began to solve this equation numerically is explained below.

$$\partial h / \partial t = f(t)$$

First, we changed the equation simply as $(h_{j+1} - h_j) / \Delta t = f(y_j, t_j)$

Second, we multiplied both sides by ∂t , and added h_j to both sides. Now, left hand side (LHS) of the equation is just h_{j+1} .

Next, we selected a Δt which is very small and every time we started add up h_j 's to the h_{j+1} . After enough iteration the value of h_{j+1} is our ∂h value.¹⁰²

In our numerical solution approach to continuum growth equation, we picked v and $\kappa=0$. Of course, they are not 0 in real but we do not have surface tension and surface diffusion terms in this thesis. We have a new approach and wrote a Matlab code that calculates F1 term in the code. We tried our code, when there is no re-emission and no surface diffusion. We saw that there is a random deposition. Since it takes too much time, lattice size of all our numerical solutions is 32×32 units. The idea how we calculate F1 terms are explained below.

First of all, we thought a substrate that is 32×32 units. Each of them has a height which is randomly generated. We calculated at a period of defined time steps, which is Δt , how many atoms can get a grid from the system. We compared all combinations of grids that if an atom can go the other place. We draw a line from two points and calculated which grids can an atom use to go from a point to a target point. We geometrically calculated all heights of over the grids. If all calculated heights are greater than grid heights, this means that atom can go the target place after being re-emitted. Ratio of all atoms which can go to target point, $h(i, j)$ to all sent atoms gives the F1 for this system. We calculate this flux for all grids, and then put all these F1 terms in the system. Now, we have new height data. We determine a number of steps, so program calculates these data until the number of steps end.

8.2 Computational Results

In Fig 8.1, we plotted rms roughness vs. time graphs for different noise values. β values for these graphs vary from 0.39 when $\eta=10$ to 0.05 when $\eta=10^{-8}$. We used $S_c=0.9$ for all these graphs. In Fig. 8.2, we plotted the same graphic for three different

noises when $S_c=0.1$. The η values that we used are 0.01, 1 and 3. The reason why we used these η terms is; when $S_c=0.9$, these η terms nearly matches with theoretical results. In Fig. 8.3, we used same η values, 0.1, but different S_c values that are 0.1, 0.5, and 0.9. We measured β values; 0.19, 0.26 and 0.30 respectively.

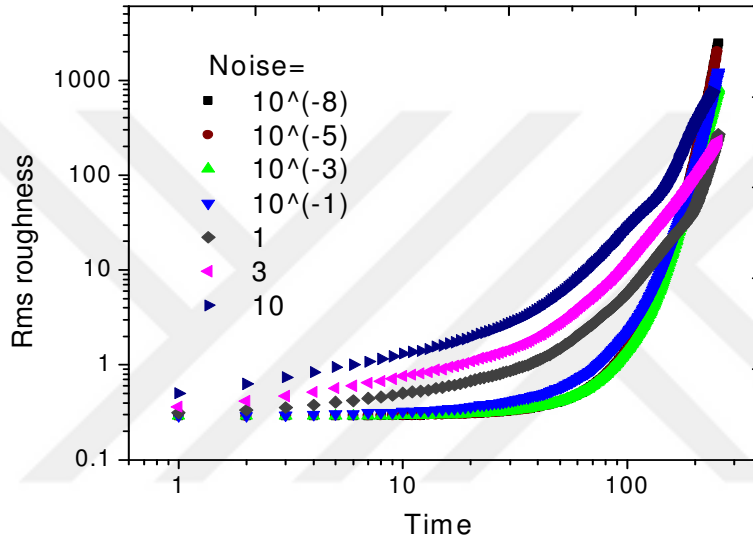


Figure 8.1: Root mean square roughness vs. time graphic when $S_c=0.9$ in first order re-emission for different noise values.

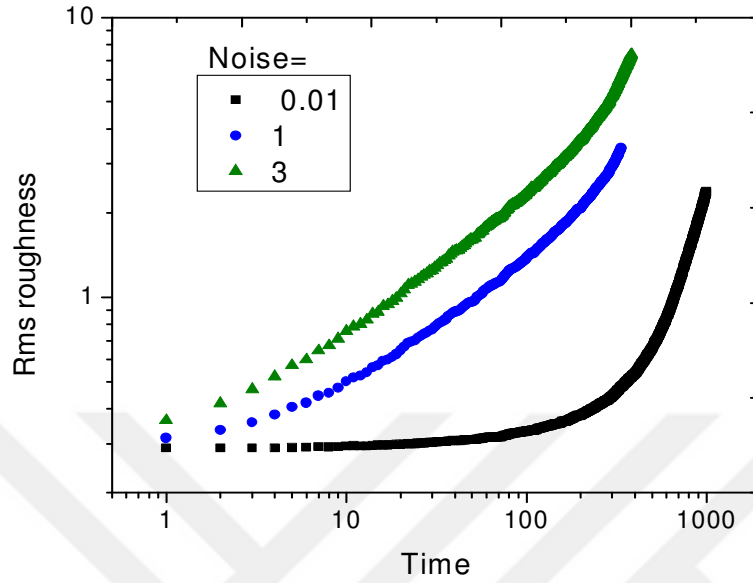


Figure 8.2: Root mean square roughness vs. time graphic when $S_c=0.1$ in first order re-emission for different noise values.

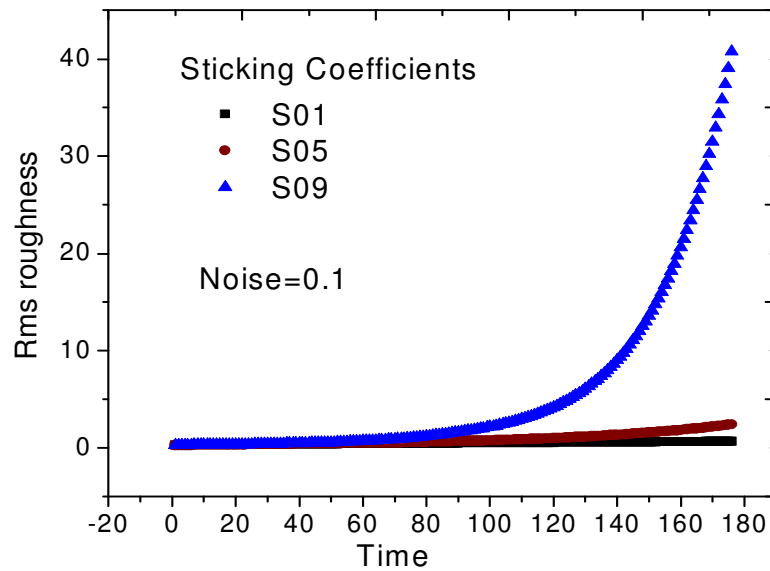


Figure 8.3: Root mean square roughness vs. time graphic when $\eta=0.1$ in first order re-emission for different S_c values.

8.3 Concluding Remarks

In this section, we focused on numerical solutions. First, we gave some backgrounds for numerical solutions. Then, we explained how we are going to apply numerical solutions to continuum growth equations. We mentioned detailed about how we approached to solutions. Last we plotted some graphs that we calculated using different η and S_c values. We first plotted root mean square roughness vs. time graph when $S_c=0.9$ in first order re-emission for different noise values. Then, we changed the S_c value as 0.1. In all these numerical simulations, we used different η and we compared how β changes with S_c and we compared if all parameters are same, how β changes with η changes. Lastly, we used same η values and compared with different S_c 's. In Fig. 8.1 and 8.2, we see that if noise increases, β increases.

9. DISCUSSION AND CONCLUSIONS

9.1 Summary and Discussion

In this chapter, we summarized the important results of this thesis and mentioned some of the key issues. We also listed some recommendations for future work at the end of this chapter.

9.1.1 Effects of Re-Emission during Thermal Evaporation Growth Using Monte Carlo Simulation Method *Without* Surface Diffusion

In chapter 5, we investigated the effects of re-emission using Monte Carlo simulations. Since we used thermal evaporation, we commanded that all the particles are coming from the top in the code. We first calculated growth exponent, β , values. We plotted β vs. S_c graphs. We realized that β is too small when S_c is less than 0.9. Although 0.9 sticking coefficient seems big, it is not big in our case. If $S_c=1$; we see that there is a random deposition. We then investigated α vs. S_c , and z vs. S_c graphs. After we analyzed height distributions, we investigated β vs. S_c in all order re-emission and saw that there is no significant difference. We compared these and some other graphs in chapter 5.3. We did all our simulations 10 times in order to find the effect of noise.

9.1.2 Effects of Re-Emission during Thermal Evaporation Growth Using Monte Carlo Simulation Method *With* Surface Diffusion

In chapter 6, we almost did same analyzes with chapter 5. However, there are some changes. First, we defined surface diffusion and diffusion rate, D/F . Then, we calculated what the correct D/F that we used in our simulations is. We calculated that at

$S_c=1$ if D/F is 350, β is equal to 0.25 which matches with theoretical results. Whole simulations that we have done are with a 350 D/F . We have done all our simulations in first order re-emission, all order re-emission and compared both of them. In chapter 6.5, we compared these results with previous chapter. Namely, we analyzed if the system has surface diffusion and not surface diffusion.

9.1.3 Surface Evolution during Growth on Patterned Substrates

In chapter 7, we did something different. We analyzed the effect of re-emission and surface diffusion, however, this time our substrate is patterned. Instead of growth exponent, we defined a new term, smoothening exponent. We used a patterned substrate and we gave the properties of substrate in this chapter. We plotted smoothening exponent, m vs. S_c for different radiuses. The most important result is that, we cannot predict S_c 's just by calculating m 's. The other important result is we can define a wavelength selection in the early deposition stages. We compared first order and all order re-emissions and saw that there is no big difference as in previous two chapters. There is a slight difference if surface diffusion exists in simulations.

9.1.4 A Numerical Solution Approach to Continuum Growth Equation

In Chapter 8, which is about numerical solutions, we numerically solved continuum equation partly and analyzed surface morphology. We first plotted root mean square roughness vs. time graph when $S_c=0.9$ in first order re-emission for different η values. We selected these η values in a wide range. Then, we lowered the S_c value. In all these numerical simulations, we used different η and we compared how β changes with S_c

and we compared if all parameters are same, how β changes with η changes. Lastly, we used same η values and compared with different S_c 's. In conclusion, we saw that if there is re-emission, our β value decreases.

9.2 Conclusion

In general, we analyzed the effects of re-emission and surface diffusion in thermal evaporation using thin film modeling. We used a code which is originally written by Jason Drotar using C++ and we modified the code for our system. Using different sticking coefficients, we measured how growth exponents change with different sticking coefficients. We saw that the both re-emission and surface diffusion has an effect, however, re-emission has more effect than surface diffusion in thermal evaporation system. Next, we used patterned substrate, and then examined the effects of re-emission and surface diffusion. At last, we plotted some rms roughness vs. S_c graphics using numerical solutions.

9.3 Recommendations for Future Work

Although thermal evaporation is an old technique, it is still a developing technique. Any theoretical or numerical study using any technique is appreciated about analyzing surface morphology. Our recommendation for improving data that we presented in this thesis is mostly about numerical solution approaches. There are some reasons for this.

The first reason is although it is slower as compared to Monte Carlo simulations, it gives better results. The reason why people did not study much about numerical

solutions is, computers are not fast enough to run numerical solution codes. Although it is not too fast, it is almost fast enough to work with these codes.

The second reason to focus on numerical solutions is, both for chemical vapor deposition and sputter deposition, people have analyzed the evolution of thin film morphology. In this thesis, we also have analyzed for thermal evaporation deposition. People can find lots of journals, books, other studies about MC simulations, and experimental techniques about analyzing surface morphology. However, there is a few studies about numerical solutions. So, we can briefly say that science needs numerical studies for measuring thin film morphology.

We recommend experimental studies for both patterned and experimental studies. All studies about comparing MC simulation results, numerical solutions and experimental studies will greatly be appreciated.

The last recommendation for future work is about developing the code for continuum equations. If we include the effect of flux terms which are higher than F_1 , and including surface tension term and surface diffusion coefficient term, it is too difficult to handle this code. It runs too slow, and it is difficult to set the right parameters such as noise term in the code. In conclusion, all types of developing the code is recommended for future work.

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APPENDIX A: Diffusion Rate

How to find surface diffusion rate, D/F , mathematically is explained below.

E_0 = The force of bond between the atom and under the atom.

E_n = The force of bond between the atom and its neighbors such as under the atom, right and left side of the atom

n = Number of nearest neighbors

Total binding energy = $E_b = (n * E_n) + E_0$

Diffusion $\sim \exp(-E_b/kT)$ and it is between 0 and 1

k : Boltzmann constant

T : Temperature

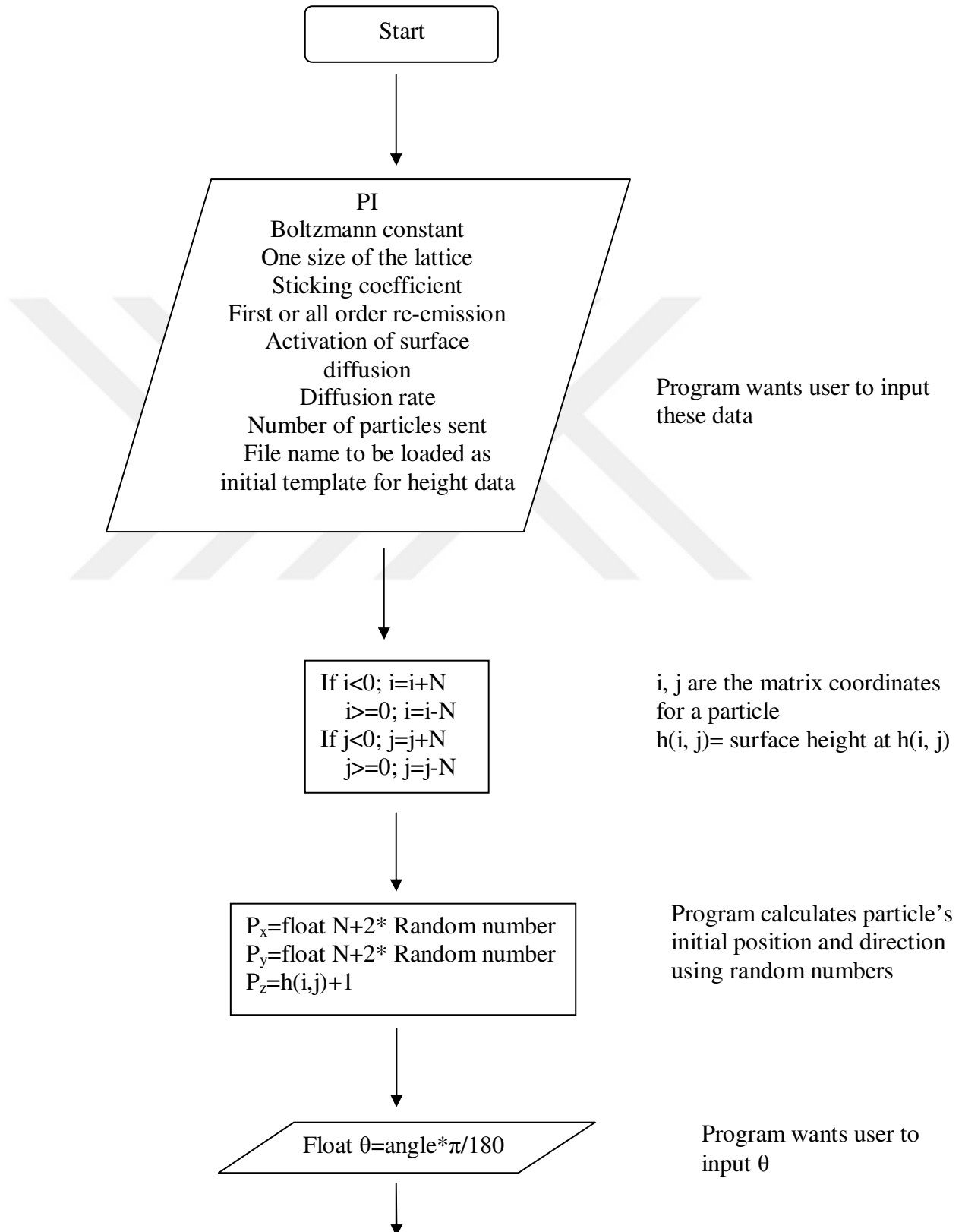
R = Random number generator and $0 < R < 1$

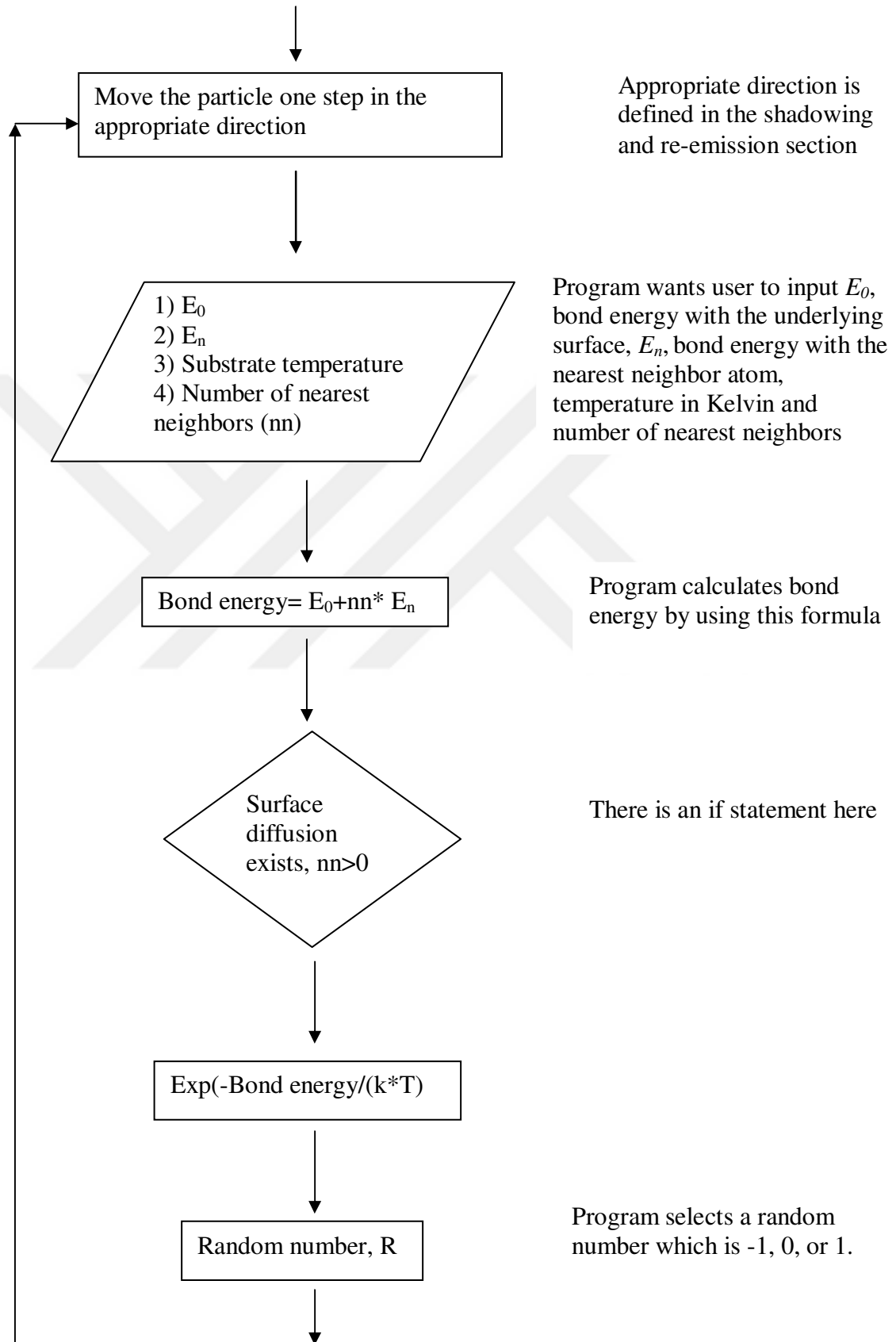
If diffusion $> R \rightarrow$ it diffuses

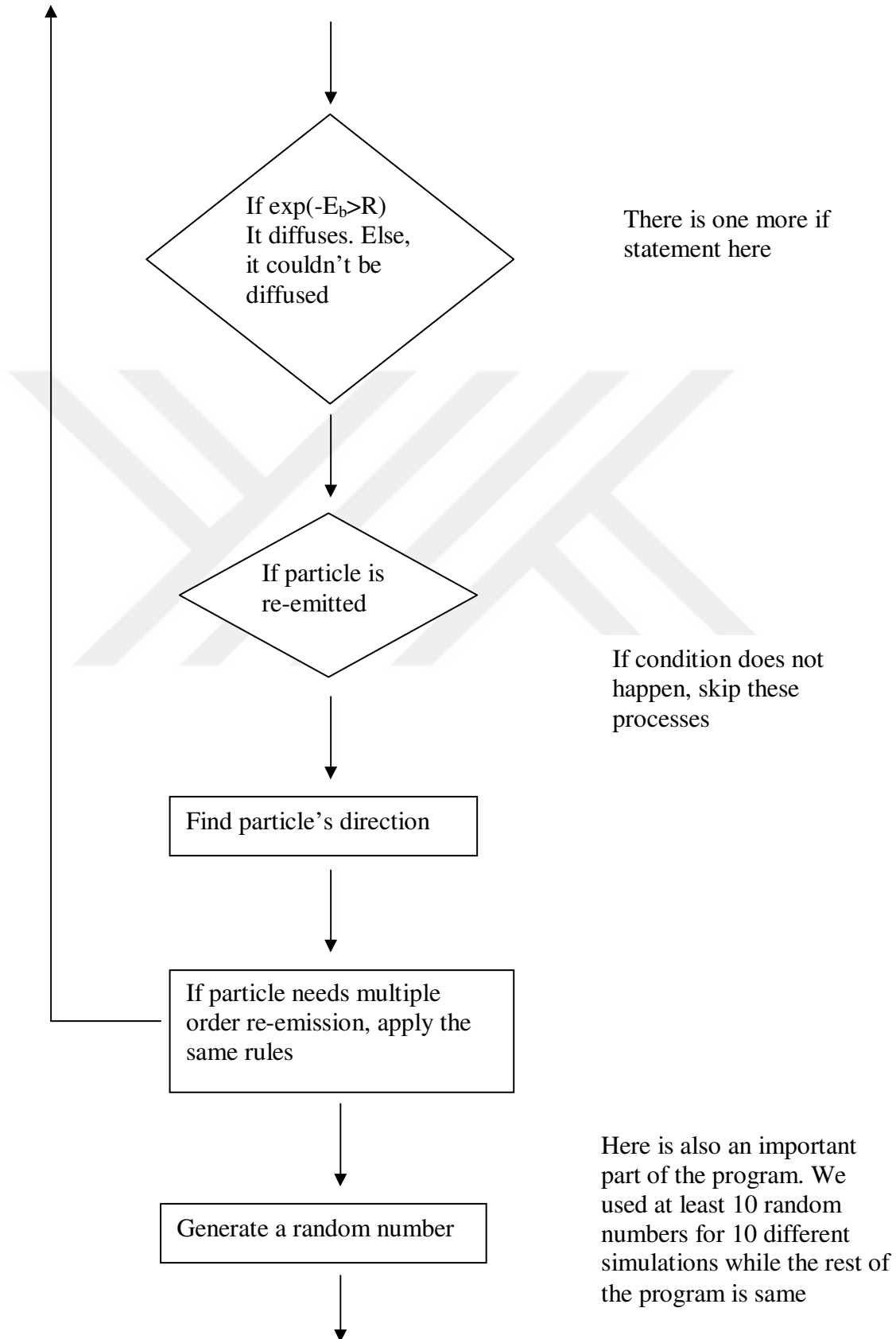
D/F = Number of atoms to be diffused = Diffusion rate

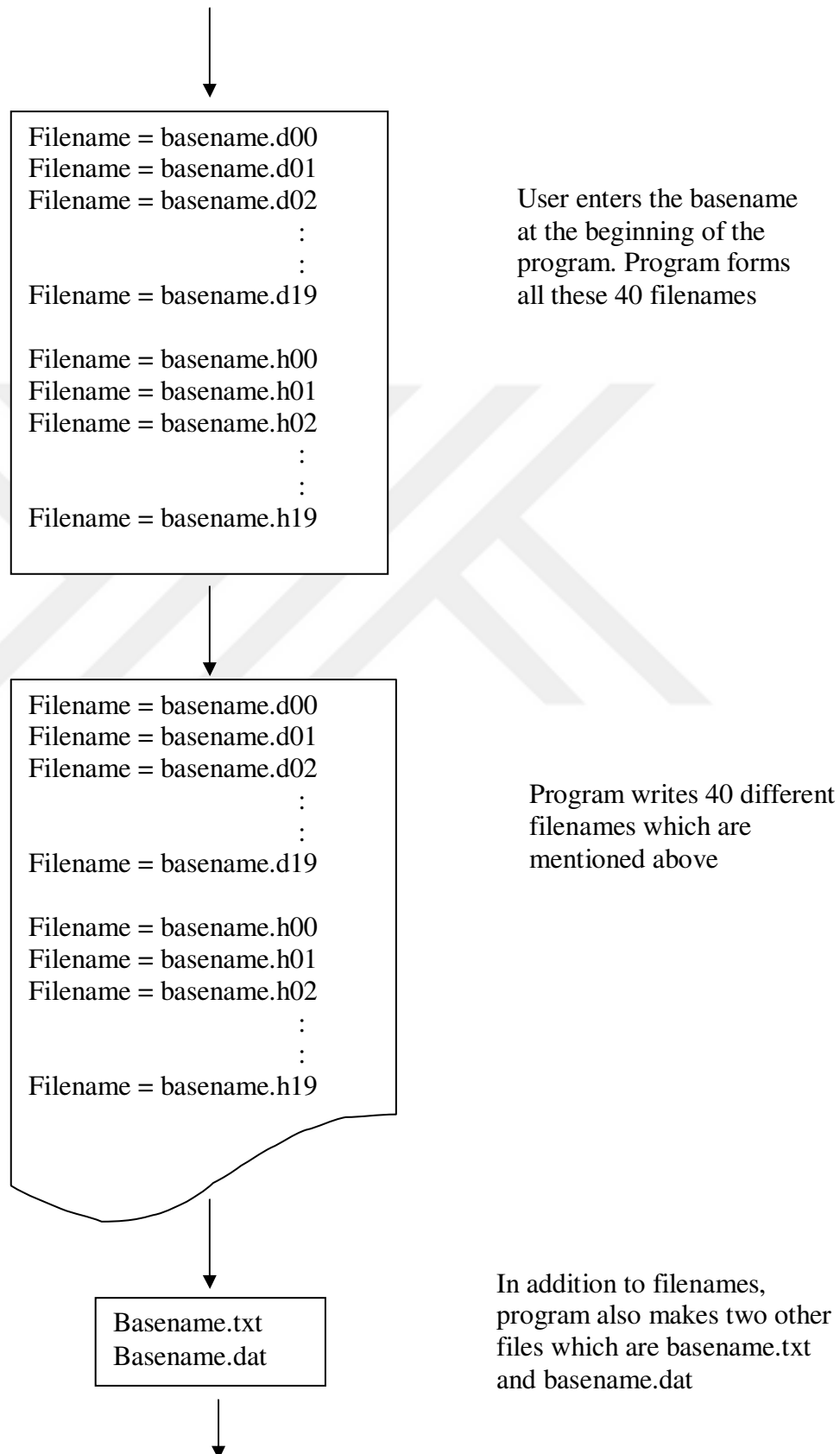
For example $D/F = 50$; but 20 atoms can be diffused since $D > R$ at 20 atoms.

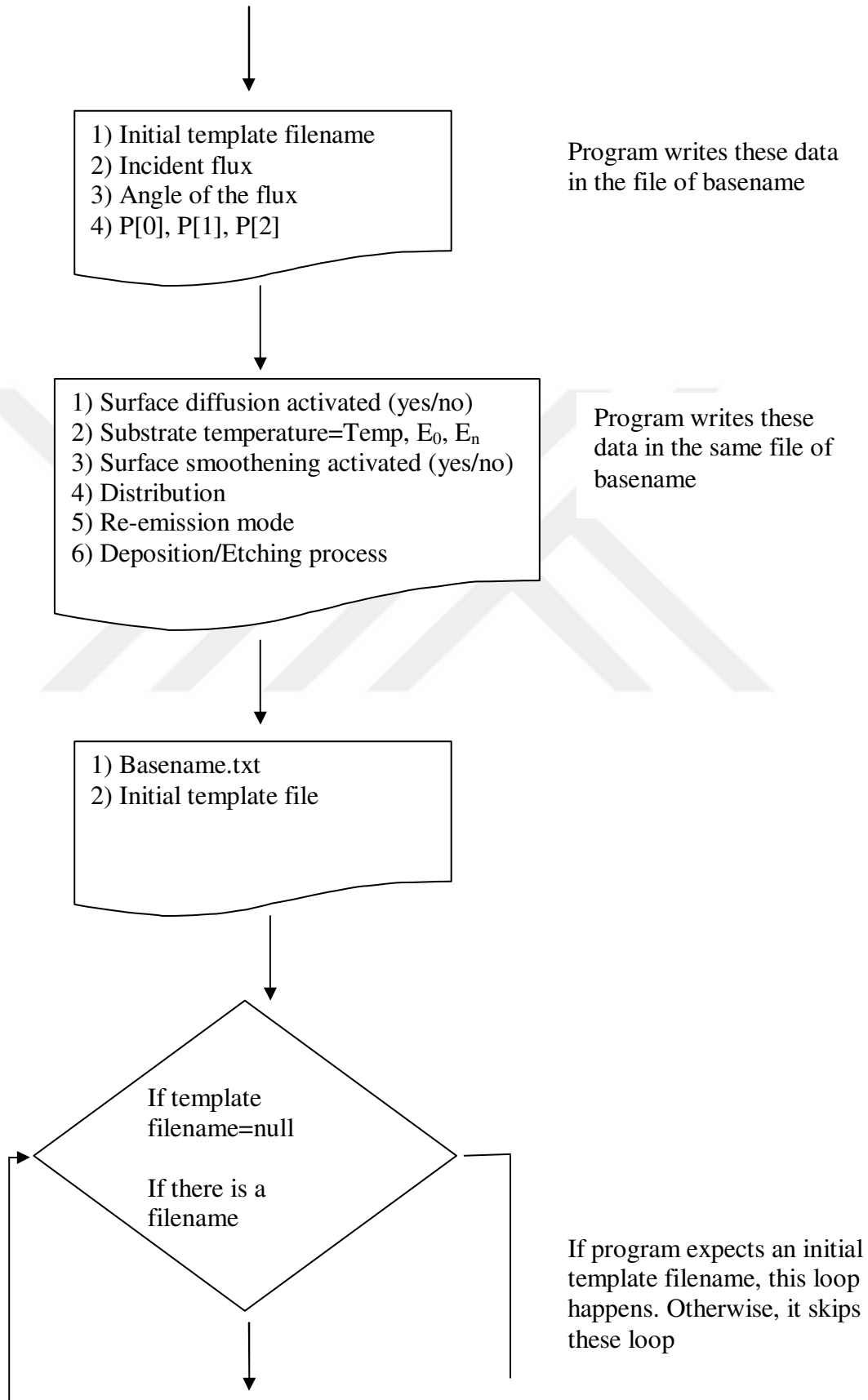
APPENDIX B: Algorithm of Monte Carlo Simulations

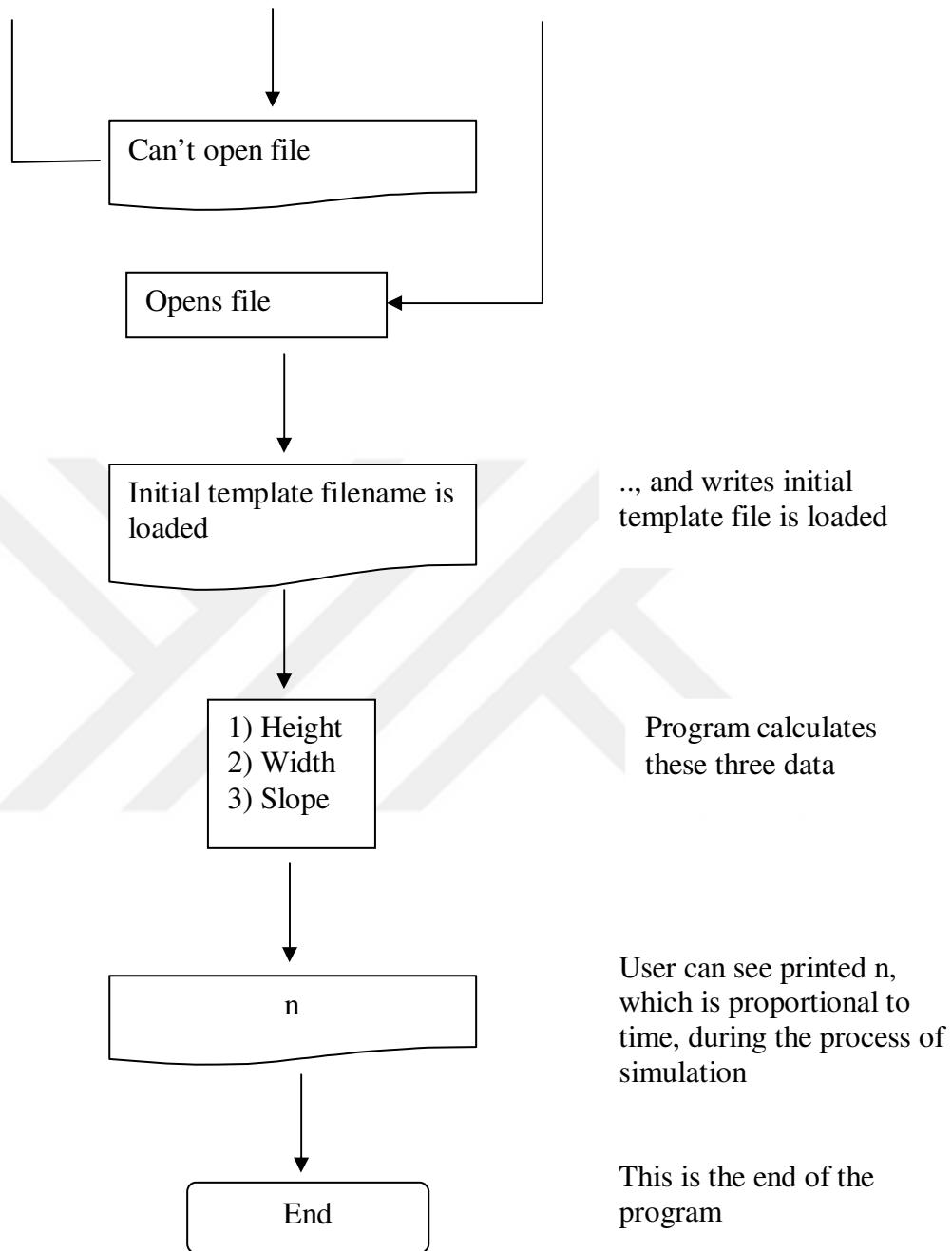












APPENDIX C: Algorithm of Numerical Simulation Methods

