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A New Random Walk Simulation Model for Study of Diffusion Behavior of Single Particle Within Two-Dimensional Space

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Abstract: Research on diffusion behaviors is of significant value in that it is closely related to transport phenomena in micro-chemistry. However, the effects of variables on diffusion are still unclear. Here, we developed and programmed a simulation methodology along with data analysis, which was capable to simulate the diffusion of a particle within two-dimensional heterogeneous space in large timescale; the effects of periodically arranged impenetrable barriers of specific shape and lateral drifting velocity on diffusion behavior were studied. As well as standard mean square displacement analysis, a new method, the appearance probability distribution method, was introduced, which revealed whether the particle tended to be present at certain positions. This article introduced the construction of the simulation model and demonstrated the validity of the model. The results showed that our model fit qualitatively well with experiments and theories. The model was proved to be an excellent potential platform for simulating the diffusion behaviors in micro-chemistry, such as the diffusion process in electrochemistry as well as nanofiltration membrane.

Key words: diffusion; random walk; simulation; mean square displacement; appearance probability distribution
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1 Introduction

The diffusion behavior of particles is a basic issue of micro-chemistry. For instance, in the field of electrochemistry, studies on the diffusion behaviors of ions in electrode[1], membrane[2] and electrolyte[3] have drawn intensive attention of many researchers. Direct experimental observation of single-particle diffusion was performed by single-particle tracking techniques with the use of fluorescence microscopy, atomic force microscopy and so forth[4-8]. These techniques are very powerful in revealing microscopic motion of molecules, but there are still some restrictions. First, the time- and space-resolution of the techniques are limited. This hinders them from revealing more detailed information of the motion. By experiment, Takimoto did not observe a plateau in diagram predicted by simulation due to the limited time-resolution of their instrument[9]. Second, the observation time range cannot be long enough to reveal the long-time diffusion behavior of particle owing to the restrictions like fluorescence photobleaching effect. Third, real diffusion process is influenced by many factors and disturbance; therefore experimental data may contain certain "noise" that conceals the true nature of diffusion. In comparison, simulation method is advantageous in that it is not restricted by experimental techniques and can simulate with rather high resolution and long time-scale. Also, parameters are readily controlled in simulation so as to study the influence of specific factors. In previous literature, simulation method was often used as an auxiliary means, but only a few aim at

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obstructed or anomalous diffusion[10-11]. For example, Niehaus[10] simulated a plane with grid-like semi-penetrable barriers and studied their effects on the diffusion of a particle. Such simulation is reasonable, but may be restricted to a certain kind without much room for further development.

Here, we developed a new and natural model to simulate single-particle diffusion process within heterogeneous two-dimensional space. The idea came from the experimental work of the research group of Murakoshi[12-15], who fabricated a microscopic metallic structure called "nanogate" and study the diffusion of molecules within the structure. We modeled the nanogate structure with an infinite two-dimensional plane containing periodic impermeable zones of specific shape, which we refer to as barriers, mimicking the metallic obstacles in the experimental work. A particle, represented by a point, takes sequential random steps among barriers and forms a trajectory. To analyze the trajectory, mean square displacement (MSD) and its first derivative with respect to time are calculated to reach the time-dependent instantaneous diffusion coefficient D. In addition to this, we also exploit a new analysis method, the appearance probability distribution (APD), which visualizes the spatial distribution of the probability of appearance to see if the particle is more likely to be located in specific region due to the environmental influence.

Diffusion can be classified to several modes according to characteristics of its MSD[8]. Four modes, namely normal, confined, anomalous and directed, can be apparently observed in our simulation. Moreover, by altering parameters, the transitions between these modes are shown clearly. Thus this new model seems to be reasonable and suitable to describe the behavior of a particle moving within heterogeneous plane. This model has been applied to simulate the molecular diffusion behaviors in nanofiltration membrane[16] as well as the membrane of lithium-ion battery[2]. Furthermore, it can also be improved on the basis of this framework by considering more effects and parameters.

2 Methods

2.1 Modeling

We designed a set of software simulating a particle stochastically diffusing within a two-dimensional space and performing data analysis as well as visualization for the simulated trajectories. The two-dimensional space is heterogeneous, with periodically arranged impenetrable regions called "barriers". Practically, we divide the infinite space into periodic "cells", each being a square, and define the side length of the cell as cell constant. A barrier is defined in each cell. The space between barriers is called plain space which a particle can pass through without disturbance (Fig. 1). A particle, represented by a point with certain x and y coordinates, is located at an initial position and start moving step by step, whose speed and direction obey certain probability distribution respectively. For all simulations in this article, speed of each step of the particle keeps constant during a simulation run, and as time of each step does not vary, step length is constant for a run. The verification of using constant speed is in Supporting Information 1. The particle moves within plain space and rebounds back when hitting the edge of a barrier. In the simplest model, the interaction between particle and barrier is set to be perfect elastic collision, obeying law of reflection.

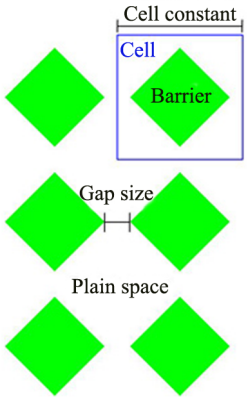

Fig. 1 Simulated heterogeneous space and basic concepts. "Cell" is the unit that is periodically repeated to cover the whole plane. "Barrier" is the region that a particle cannot step into. "Plain Space" is the space other than barrier. "Gap Size" is the spacing between two nearest barriers.

To set up a simulation, parameters need to be specified. Unit of parameters of length dimension, like cell constant, can be micrometer or nanometer; unit of parameters of time dimension can be microsecond or nanosecond. What is significant is not the very unit we use for parameters but the relative ratio of their value when represented in the same unit. Parameters can be interpreted as having any specific unit, depending on research. As this article aims at obtaining general information of diffusion, all units of parameters here are omitted. The validity of this model is examined in Supporting Information 2.

2.2 Simulation

First, set up the following parameters: simulation times (how many simulations in a single run), cell constant (side length of the cell), barrier parameters (coordinates of the vertices of the barrier), initial state (initial position of the particle and cell), velocity parameters (speed distribution type and corresponding parameters, lateral drifting velocity which will be described later), step time (time duration of one step), total number of steps in a single run. Then the simulation begins; the particle moves step by step within the plain space, the speed of each step obeys the chosen distribution and the direction being uniformly distributed in 360° . Despite this random velocity, there can be also additional lateral drifting velocity whose direction is fixed, say, along x direction, mimicking the effect of external field like electric field. The position coordinates of the particle after every step are recorded for analysis. As the simulation finished, a trajectory is obtained by linking position points in time sequence. Data analyses described below are performed. Typically simulations are performed in sets. In each set, several tens of simulations with the same setting are carried out and analyzed in a row, and the consequent data are averaged to get more reliable results.

2.3 MSD Analysis

MSD of step t is calculated by adding up the squared displacement of every pair of position coordinates whose step interval is t. It is a function of time (or step) t and given by

$$\mathrm{MSD}(t) = \frac{1}{N - t + 1}\sum_{k=0}^{N-t} |d_{k+t} - d_k|^2 \quad (1 \leqslant t \leqslant N) \qquad (1)$$

in which N is the total step number, d_k represents the position coordinate (a position vector) of the particle at step k (d_0 represents the initial position) and $|d_{k+t} - d_k|$ represents the displacement of the particle from step k to step $k+t$. MSD_x and MSD_y are given by

$$\mathrm{MSD}_x(t) = \frac{1}{N - t + 1}\sum_{k=0}^{N-t} |d_{x,k+t} - d_{x,k}|^2 \quad (1 \leqslant t \leqslant N), \qquad (2)$$

$$\mathrm{MSD}_y(t) = \frac{1}{N - t + 1}\sum_{k=0}^{N-t} |d_{y,k+t} - d_{y,k}|^2 \quad (1 \leqslant t \leqslant N), \qquad (3)$$

in which $d_{x,k}$ and $d_{y,k}$ are the x and y component of the position vector d_k respectively. Evidently the following relation always holds:

$$\mathrm{MSD}(t) = \mathrm{MSD}_x(t) + \mathrm{MSD}_y(t). \qquad (4)$$

MSD is an index of how far a particle travels in a certain time: the larger MSD is, the further the particle travels. The first derivative of MSD with respect to t is an index of how fast a particle travels at a specific moment. Numerically, it is calculated by

$$\frac{\mathrm{dMSD}}{\mathrm{d}t}\Big|_{t=t_0} = \frac{\mathrm{MSD}(t_0+1) - \mathrm{MSD}(t_0-1)}{2}. \qquad (5)$$

For normal diffusion within two-dimensional space with no barrier, $\frac{\mathrm{dMSD}}{\mathrm{d}t} = \frac{\mathrm{MSD}(t)}{t} = 4D$ in which D is diffusion coefficient. $\frac{\mathrm{MSD}_x(t)}{t} = 2D_x$ and $\frac{\mathrm{MSD}_y(t)}{t} = 2D_y$ can be defined similarly as for one-dimensional diffusion. For other modes of diffusion, $\frac{\mathrm{MSD}_x(t)}{t}$ may not be a constant; in this case we still define

$$4D(t) = \frac{\mathrm{dMSD}}{\mathrm{d}t}, \qquad (6)$$

in which $D(t)$ is dependent on t, but can still reflect the diffusion rate of a particle. The $D(t)$ defined in this way can be called instantaneous diffusion coefficient.

2.4 APD Analysis

This analysis aims at revealing the probability of appearance of the particle at different positions in

the space during a simulation run. Precisely, first, all position coordinates are normalized to be relative to a single cell exploiting the periodic nature of the space. Second, the cell is transformed into an $n \times n$ grid, and the number of position coordinates located in each unit of the grid are counted and normalized to get the appearance probability. Third, the probability distribution is shown on the screen with different color representing different probability value. This process is programmed and can be performed automatically.

3 Results and Discussion

3.1 General Influence of Barrier and Comparison with Experiment

To have an overview on the influence of barrier, a two-dimensional space configuration was set up as following. Barriers were set as diamonds resulting from 45° rotation of a square, located at the center of each cell (as Fig. 1 shows). Cell constant was set to be 10, random velocity to be 1, and step time to be 1. Gap size was altered to examine how the size of barrier affects the diffusion behavior. Two control sets were also carried out: a free diffusion set with no barrier, and a confined diffusion set in which gap was 0, i.e. the particle was confined in a single compartment. For each set, 30 independent trajectories of 300 000 steps were simulated.

MSD-t curves of these sets are shown in Fig. 2. In a low-resolution graph, all curves seem to be straight lines, but closer examination of the $t < 100$ region reveals that the curves are not straight except that of the free diffusion set. The curve of greater gap/cell ratio increases more rapidly with time due to the less confinement of the barriers. Least square fitted equation of $t = 1 \sim 1000$ of the free set is MSD = 0.99493t - 0.28809 ($R^2 = 1$). Theoretically, MSD = $v^2\ \tau\ t$ (see Supporting Information 3) where v is step speed, $v = 1$, and τ is step time, $\tau = 1$, so in this case MSD = t. Simulation result fits well with theoretical result. The confined diffusion set is different, though. MSD increases as t increases when $t < 100$ and goes to a steady value, 16.7, thereafter. As the particle cannot escape the initial compartment, its

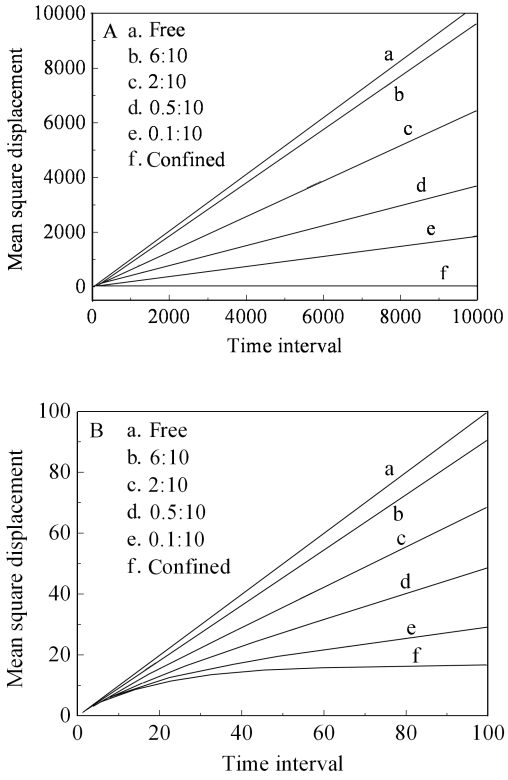

Fig. 2 Mean square displacement curves of different gap/cell ratio (A) and its detail of the short-time region (B).

displacement must be less than the longest distance between two positions in the compartment, so it cannot keep on increasing with time.

The steady value of long-time MSD of confined diffusion can be derived theoretically. MSD(t) is the mean value of squared displacement between two positions $r_{(k)}$ and $r_{(k+t)}$, every pair of which actually represents a segment of the whole trajectory. The mean is calculated over a great amount of such segments; take this simulation as an example, MSD (1000) is the mean over (300 000-1000+1) × 30 = 8 970 030 segments. With such amount of samples, rules of probability come into effect. For a segment long enough (when t is large), its two end points can be located at any position in the compartment with equal probability. Consider the x component of the end points, x_i and x_f, both having the probability density $1/a$ to be located at any x in an $a \times a$ compartment. MSD_x can be calculated by

$$MSD_x = \langle (x_f - x_i)^2 \rangle = \int_0^a \int_0^a (x_f - x_i)^2 \frac{dx_f}{a} \frac{dx_i}{a} = \frac{a^2}{6}. \quad (7)$$

The situation is identical for y component. Then, $\mathrm{MSD} = \mathrm{MSD}_x + \mathrm{MSD}_y = \frac{a^3}{3}$. The case above in which $a = \frac{10}{2}\sqrt{2} = 5\sqrt{2}$ gives MSD = 16.7. This is exactly the same as the simulation result. Other sets with different gap/cell ratio ranging from 0.1/10 to 6/10 are between the two extreme controls, and the larger the ratio is, the closer the set is to the free one. This shows that anomalous diffusion is a transition mode between two extremes: normal and confined diffusion.

The characteristics of MSD can be further revealed by log-log curves of instantaneous diffusion coefficient, $D(t)$, versus t as shown in Fig. 3. The curve of free diffusion is a horizontal line, for D keeps constant during the diffusion. The curve of confined diffusion drops sharply to about 0 at around t = 100. Curves of other diffusion in space with barriers follow the same trend that they decrease, first "accelerate" then "decelerate" and finally go to a steady value, which showing confinement effect of the barriers. This phenomenon is consistent qualitatively with experiment[9]. Such decrease is due to the confinement of barriers to the diffusing particle. In short time, the particle moves mainly within a single cell behaving close to free diffusion with mildly decreasing D (micro-D). As time increases, the particle diffuses among cells, being significantly affected by the confinement effect of barriers, with obvious decreasing D. The process being a Markov chain, diffusion from one cell to another should be independent and the average time be characterized by the setting of simulation parameters. Thus, diffusion from cell to cell would behave like free diffusion, with a smaller constant D at long-time scale, defined as macro-D. The phenomenon that at long-time scale confined diffusion would behave like free diffusion has been visualized by Niehaus et al[10].

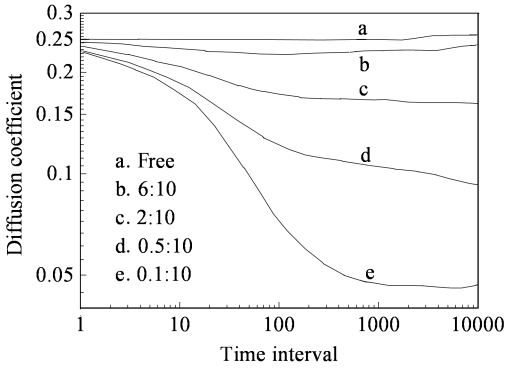

Fig. 3 Diffusion coefficient curves of different gap/cell ratio.

APD analysis was also performed for all simulation runs. The distributions are all homogeneous (Fig. 4), i.e. the particle appears at arbitrary position in plain space with equal probability, which shows that the confinement of barriers does not create a bias in the presence position of a diffusing particle. This seems to contradict the research of Ritchie et al.[11], which showed that the particle tended to appear in the central region of compartment. In fact, the phenomenon in the research of Ritchie et al. is due to the effect of "frame time", that is, when using large "frame time", the "recorded" positions tends to be in the central region, but if counting "unrecorded" positions as well, the total positions are actually distributed homogeneously over the plain space.

3.2 Influence of Compartment Size

To investigate the influence of particle's speed r elative to compartment size, the space configura tion was set up as following. Barriers were also set to be diamonds resulting from 45° rotation of a square, located at the center of each cell. The ratio of gap to cell constant was set to be 1:100, a small value to show strong confinement effect. Step time was set to be 1, random velocity to be 1, and cell constant (along with size of barriers) was altered to

Fig. 4 Appearance probability distribution graph of gap/cell =1/10 (APD of other gap/cell ratio are similar to this).

change the size of compartment. For each set, 30 independent trajectories of 300 000 steps were simulated.

MSD-*t* and *D*-*t* curves of these sets are shown in Fig. 5. Previous researches[10, 17] have revealed that such *D*-*t* curves have a plateau region when *t* is small, and this can be readily seen in the simulated curves. As the size of compartment increases, the plateau expands to larger *t*. This can be explained by the confinement effect of barriers. Larger compartment provides more space for the particle to diffuse freely, and thus the particle behaves like a free one in longer time scope, resulting in more extended plateau on the curve. Also macro-*D* increases as the ratio of speed to cell constant decreases, owing to the weakened confinement of barriers as well. The distribution of micro-*D* and macro-*D* of each set is just the same as experiment result[17].

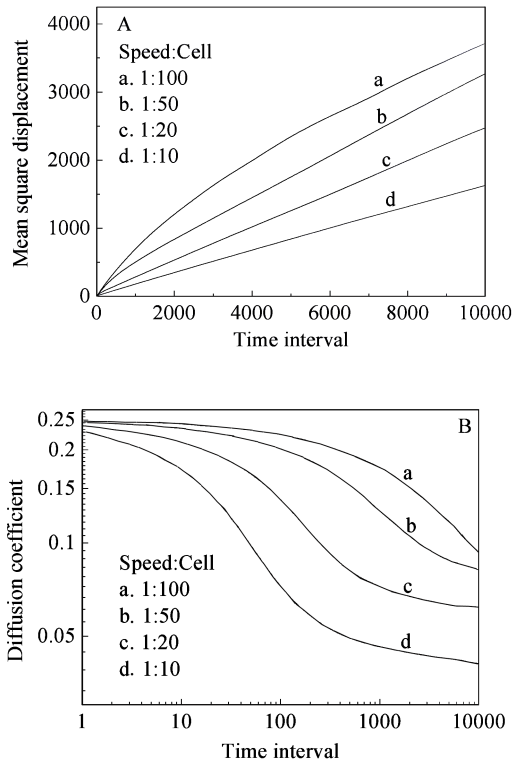

Fig. 5 Mean square displacement curves (A) and diffusion coefficient curves (B) of different compartment size (speed/cell ratio).

3.3 Influence of Available Space within Compartment

Not only compartment size but available space within compartment as well can affect the diffusion behavior. To investigate the influence, barrier with special shape was designed as shown in Fig. 6A. It was a combination of a square and a cross (the area of the cross can be neglected comparing to the area of the square); the cross was to control the gap size of a compartment while the square was to control the available space within compartment. The cross was fixed and the size of square was altered to investigate the influence. Other parameters were: gap = 1, cell constant = 10, random velocity = 1, step time = 1; 10 independent trajectories of 300 000 steps were simulated for each set.

The *D*-*t* curves are shown in Fig. 6B. It turns out that as the size of the square part of barrier increases (that is, the available space shrinks), the micro-*D* decreases but the macro-*D* increases; the "slope" of the curve becomes flatter. It is because shrank space within compartment constrains the particle's diffusion and thus causes micro-*D* to decrease, but it also urges the particle to move out of the original compartment and diffuse among compartments, causing macro-*D* to increase. For the extreme set that the square part totally covers the cross part of barrier, there is no distinguishable compartment, and the curve becomes horizontal after few steps. This is very much like the curve of a normal diffusion. This has important implication. It seems, strictly speaking, that anomalous diffusion mode may be directly due to the formation of compartment by barriers rather than the interaction itself between barrier and particle. As in this case, most part of the two-dimensional space is covered with barriers and the interaction is so strong that it causes the *D* to drop to 0.134 (in comparison, the *D* of real free diffusion is 0.25), but its shape appears to be of the normal type. Experimentally observed normal diffusion mode may not necessarily indicate a free and undisturbed motion but possibly with

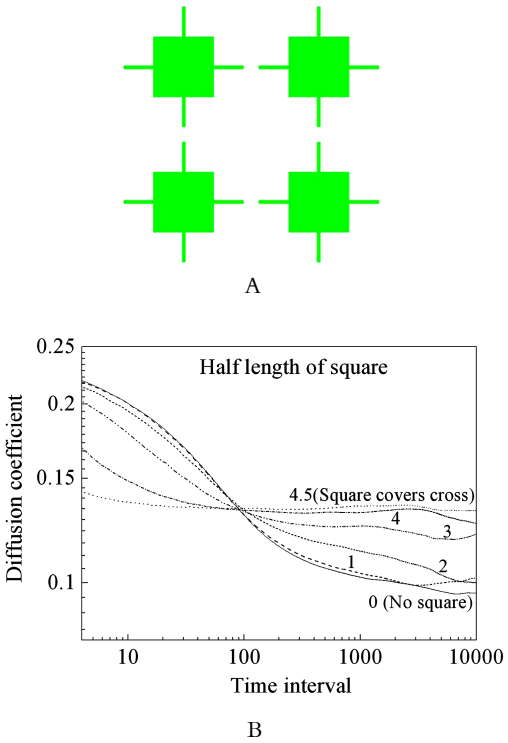

Fig. 6 Schematic diagram of "cross + square" barriers (A) and diffusion coefficient curves of different available space of compartment (B).

non-compartmental barriers.

3.4 Influence of Lateral Drifting Velocity

In experiment or application, external force along a certain direction is often exerted so that the particle would be biased along a certain direction. This force was simulate d here by adding a lateral drifting velocity to the random velocity. The basic settings of parameters were the same as Series 1 except that a lateral drifting velocity in x direction, v_x = 0.1, was added to every step.

The MSD$_y$-t curves (Fig. 7A) are just like those cases without lateral velocity because the movement of the particle along y direction is unaffected by the lateral velocity along x direction. However, the MSD$_x$-t curves (Fig. 7B) are very different from those cases without lateral velocity. For the set with no barrier, MSD$_x$ is a quadratic function of t and given by $\left\langle x_n^2 \right\rangle = \frac{\delta^2}{2} n + \varepsilon^2 n^2$ (see Supporting Information 3). The MSD$_x$-t curves of other sets with barriers are similar to the free case but correspondent MSD$_x$ is smaller. The D-t curves of barrier-presented sets (Fig. 8), however, are different from that of the free case. There are two factors affecting the particle: the confinement of the barriers constrains its movement while the lateral velocity urges it to move faster and further. At the beginning there is a "competition" between the two effects. For the case with strong confinement effect (gap = 0.1), D decreases

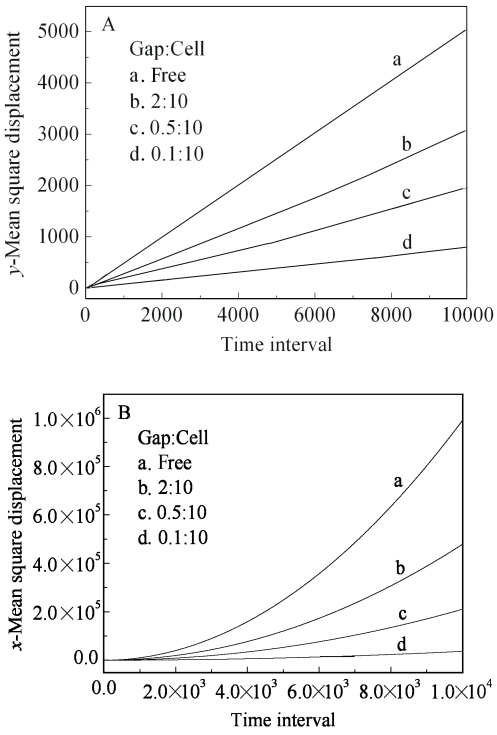

Fig. 7 Mean square displacement in y direction (A) and x direction (B) of different gap/cell ratio with lateral velocity along x direction.

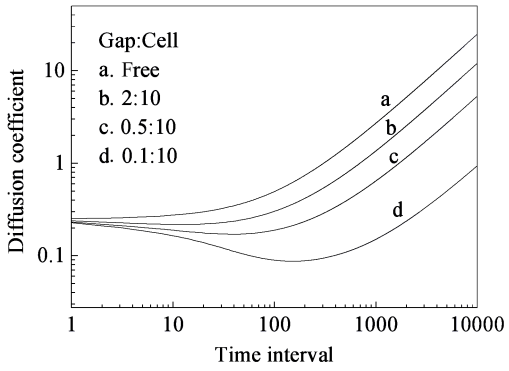

Fig. 8 Diffusion coefficient curves of different gap/cell ratio with lateral velocity along x direction.

as t increases when t is less than about 150. However, the confinement of the barriers is limited due to the Markov nature of the process whereas the effect of lateral velocity is accumulated, so in the long run the lateral velocity effect dominates the particle diffusion and D increases steadily. This phenomenon can be tested by experiment.

APD analysis shows that the simultaneous existence of lateral drifting velocity and barrier has apparent influence on the appearance probability of the particle. Fig. 9a is the APD graph when lateral drifting velocity present alone. Red points representing high appearance probability are uniformly distributed. Adding barriers (Fig. 9b ~ f), however, the distribution is no more uniform. The probability moves as a whole towards x direction (right hand side in the graphs) which is the direction of the lateral velocity, and accumulates near the right side of compartment. This trend is strengthened as the gap between barriers narrows, and the probability moves from left and central region of the compartment towards the gap and accumulates around the gap.

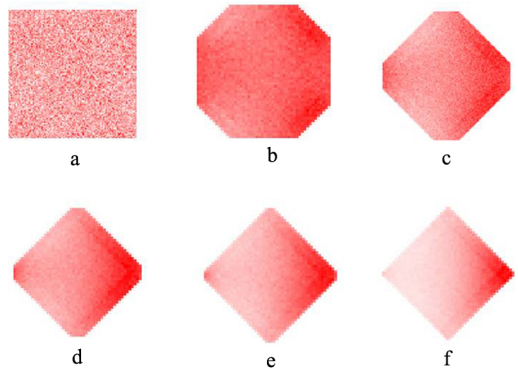

Fig. 9 Appearance probability distribution graphs of different gap/cell ratio with lateral velocity along x direction. a. no barrier; b. gap/cell = 5/10; c. gap/cell = 2/10; d. gap/cell = 1/10; e. gap/cell = 0.5/10; f. gap/cell = 0.1/10.

4 Conclusions

As we have shown, results from our model conform well to statistical theories and experiments. For normal diffusion (with no barrier presented), total confined motion (the particle is confined in a single compartment) and directed diffusion with no barrier, simulated MSD fitted quantitatively to the prediction of statistical theory; for anomalous diffusion (with restriction by barriers), simulated MSD fitted qualitatively well with experimental observations. These comparisons demonstrate that our model is justified to simulate systems that can be described by random walk. With this model we predict that given definite inter-compartment distance and passage size, the decrease in available area in compartment leaded to decline in micro-D plateau and increase in macro-D plateau, making D-t curve flatter; in extreme case that compartments became undistinguishable, D-t curve appeared to be horizontal. This observation implied that the anomalous diffusion mode is not simply due to the restriction of barriers but rather to the formation of compartment by barriers. APD analysis shows that the presence of barriers or lateral directed force alone does not create bias on the location of the particle, whereas the combination of both does. These predictions can be tested experimentally in further studies. By now the model is still simple and rough, but the fundamental framework has been built. The model can be further developed by adding detailed interactions between particle and barrier, considering the influence of solvent, etc. With these improvements, the model may become a fine tool for simulation of more systems.

Supporting Information Available

The supporting information is available free of charge via the internet at http://electrochem.xmu.edu.cn

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基于随机行走的二维空间扩散模拟研究

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摘要: 分子的扩散行为是微观化学的重要研究领域. 影响扩散行为的因素很多,但是目前各个因素的具体影响效果还不明确. 作者基于随机行走理论建立了分子在二维空间的扩散模型,依据此模型自主开发了模拟软件以及数据分析系统,并利用该模拟软件系统研究了势垒、横向速度等因素对扩散行为的影响,验证了该模型的可靠性,证明根据该模型可以得到和实验、理论相吻合的结果. 该软件有望成为模拟微观化学扩散行为的潜在平台,如电化学以及膜过滤过程中的扩散.

关键词: 扩散; 随机行走; 模拟; 均方位移; 表观概率分布