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# Hysteresis phenomena in CO catalytic oxidation system in the presence of inhomogeneities of the catalyst surface

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The hysteresis phenomena in the CO catalytic oxidation system are studied by Monte Carlo simulation in the presence of the inhomogeneities of the catalyst surface. We show that the O-passivated state is destroyed due to the inhomogeneities of the surface, in contrast to the classical Ziff-Gulari-Barshad model. The defects on the surface have a significant effect on the hysteresis transition points. Most importantly, the supercritical nucleation and growth of the O adatom island during the transition from a low reactivity to a high reactivity states are closely related to the inhomogeneities of the catalyst surface. It is shown that the width of the hysteresis loop shrinks as the scan rate $\beta_{\mathrm{CO}}$ of $y_{\mathrm{CO}}$ (the fraction of CO in gas phase) decreases, but there exists a finite width of the hysteresis loop even if $\beta_{\mathrm{CO}}$ becomes infinitely small. On the other hand, the width of the hysteresis loop decreases with decreasing the diffusion rate, and even the hysteresis loop may disappear for a slow diffusion. These simulation results are in good consistency with the previous relevant experimental results.

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## I. INTRODUCTION

The study of the dynamical behavior of open nonequilibrium systems has attracted a great deal of interest over the last several decades and has grown to a productive research area, since they possess an important theoretical value and wide-ranging applications in many branches of physics, chemistry, biology, and even sociology [1,2]. In these systems, the dynamical behavior has shown a real wealth of complexity about kinetic oscillation, chaos, spatiotemporal pattern formation, or hysteresis phenomenon [3,4]. Compared to the complicated three-dimensional chemical reaction systems such as the Belousov-Zhabotinskii reaction and Bray-Liebhafsky reaction [3,5], much attention has been paid in recent years to some heterogeneous catalytic reaction systems such as the CO and NO catalytic oxidation systems on various metal surfaces [6]. Many research results about the complicated dynamical behavior, such as the complicated temporal and spatiotemporal oscillation, in the CO catalytic oxidation system on platinum surface were reported [6–12].

Some experimental works have pointed out that there are hysteresis phenomena in CO catalytic oxidation on platinum surface [10–14]. On the macroscopic scale, the catalytic reaction system exhibits two stable steady states, i.e., one is a low reactive rate state, where CO molecules are predominated on the catalyst surface, and another steady state is high reactive state, where the catalyst surface is covered by lots of oxygen adatoms. A transition between these two steady states becomes possible if the external control parameter, such as the CO partial pressure, is varied back and forth as other external parameters are kept unchanged. A hysteresis loop may appear as a result of the transition between the two stable steady states.

Many theoretical models on hysteresis phenomena are based on the nonlinear partial differential equation or ordinary differential equation following the Langmuir-Hinshelwood (LH) mechanism [11–13,15,16]. The main problem of these models is to discuss whether a region of bistability between the two stable steady states exists. Based on the ordinary differential equation following the LH reaction steps, one can easily get a simple S-shaped curve which includes three steady states. One of the steady states is unstable and just mathematically relevant, while the two stable steady states represent the high reactive state and low reactive state, corresponding to the O-covered state and CO-covered state, respectively. If the diffusion of adsorbed CO is taken into account, a diffusion term should be added to the ordinary differential equation of the homogeneous model with the LH mechanism. Berdau and coauthors [13] have pointed out that such a deterministic mesoscopic model is unable to explain the nucleation of a new state.

On the other hand, some lattice-gas models were proposed to investigate many details of the catalytic oxidation system, such as the diffusion, desorption, and the interaction of the adsorbed species [17–22]. Evans and coauthors [23] provided a lattice-gas model to discuss the effect of interfacial CO diffusion between the different nanoscale Pt facets on the dynamical behavior of the reaction system.

In this paper, we propose a lattice-gas model to study the effect of the inhomogeneities of the catalytic surface on the hysteresis behaviors by considering the different adsorption of oxygen and desorption of CO. A hysteresis loop is exhibited in our model and some details in the experimental processes are reproduced in our simulation. We find that the inhomogeneities will have an important effect on the formation of a new phase at the transition between states of high reactivity and low reactivity. The present paper is organized as follows: Section II is devoted to the model and simulation algorithm in detail. In Sec. III, the simulation results for elucidating the influences of the external control parameters on the hysteresis phenomena are obtained and discussed. In Sec. IV, we give some discussions about the mechanism of the hysteresis phenomena. Finally, a main conclusion is given in Sec. V.

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## II. MODEL AND SIMULATION

In principle, the defects or inhomogeneities could affect the reaction on the surface in various subtle ways [13,24–27]. The inhomogeneities of the catalyst surface may be shown from two different aspects, i.e., the desorption of adsorbed CO or the adsorption of the oxygen molecule on the surface are not homogeneous. Therefore, we assume two types of defects due to the different dynamical behaviors of the two species. On the type-1 defect, the desorption of adsorbed CO can be easily realized because the binding energy of adsorbed CO on this type of defect is lower than that on the other unperturbed sites. On the type-2 defect, the adsorption of the oxygen molecule is inhibited, and at the same time, the desorption of adsorbed CO becomes more difficult than that on the surrounding unperturbed lattice sites. On the other hand, the adsorbed CO can diffuse rapidly on the catalyst surface and has a significant effect on the dynamical behavior [6,28], which is therefore included in our model. Furthermore, the catalytic chemical reaction cycle includes the CO and $O_2$ adsorption, the CO desorption, and LH reaction steps, leading to the formation of $CO_2$.

To simulate the above steps, we define some dimensionless parameters for the different dynamical processes as follows: The diffusion of adsorbed CO occurs with the probability $1-p_{chem}$, where $p_{chem}$ is the relative probability of chemical processes, including the CO desorption and adsorption-reaction steps of the two species with respect to the diffusion of adsorbed CO. Since the diffusion is faster than the chemical processes, $p_{chem} \leqslant 1$. The relative rate of the adsorption-reaction steps to the desorption of adsorbed CO is $p_{ad}$. Furthermore, the desorption probability $p_{des1}$ on the type-1 defect is much more larger than the desorption probability $p_{des}$ on the unperturbed lattice sites. However, the desorption probability $p_{des2}$ on the type-2 defect is lower than $p_{des}$. In addition, the sticking coefficients for oxygen and carbon monoxide on lattice sites are assumed to be $p_a$ and $p_b$, respectively.

The Monte Carlo (MC) algorithm for simulating the reaction kinetics on the catalytic lattice is as follows.

(1) A random number $\rho$ ($\rho \leqslant 1$) is generated. If $\rho > p_{chem}$, a diffusion trial of adsorbed CO is executed [item (2)], otherwise if $\rho \leqslant p_{chem}$, a desorption or adsorption-reaction trial is realized [items (3) and (4)].

(2) For the diffusion of adsorbed CO, a site is chosen randomly. If the site is vacant or occupied by O atom, the trial ends. Otherwise, a CO molecule located in this site tries to diffuse, and then an adjacent site is randomly selected. If the adjacent site is vacant, the CO molecule exchanges with it, and reacts with one nearest-neighbor O atom randomly if the adjacent site is an O atom.

(3) The chemical process occurring on the surface includes the desorption of adsorbed CO and the adsorption reaction of two species. A random number $\rho_1$ ($\rho_1 \leqslant 1$) is generated. If $\rho_1 > p_{ad}$, a CO desorption is performed and the desorption probability for CO on different lattice sites is different due to the inhomogeneities of the lattice surface. A site is then selected randomly. If the site is vacant or occupied by an O atom, the trial ends. Otherwise, a CO molecule located in this site may desorb. A random number $\rho_2$ ($\rho_2 \leqslant 1$) is generated. If $\rho_2$ is less than $p_{des}$ when the lattice site is an unperturbed one (when the type-1 or type-2 defect is selected, the desorption probability $p_{des}$ will be replaced by $p_{des1}$ or $p_{des2}$, respectively), the CO molecule desorbs.

(4) If $\rho_1 \leqslant p_{ad}$, an adsorption-reaction attempt is performed. An adsorption-reaction attempt contains several steps.

(i) A random number $\rho_3$ ($\rho_3 \leqslant 1$) is generated. If $\rho_3 \leqslant y_{co}$ (the fraction of CO in gas phase), a CO molecule is chosen to try to adsorb on the lattice surface, otherwise an oxygen molecule is chosen.

(ii) If a CO molecule is chosen, a lattice site is selected randomly. If the site is occupied, the trial ends. Otherwise, a random number $\rho_4$ is generated. If $\rho_4 \leqslant p_b$, the trial is accepted and CO is adsorbed, and if there are adsorbed oxygen atoms on the four nearest-neighbor sites, CO reacts randomly with one of the oxygen atoms immediately and the formed $CO_2$ desorbs at once and leaves two vacant sites.

(iii) If an oxygen molecule is chosen, a pair of two nearest-neighbor (NN) sites is selected randomly on the lattice surface. If one of the two sites is occupied or one site of the pair is a type-2 defect, the trial ends. Otherwise, a random number $\rho_5$ is generated. If $\rho_5 \leqslant p_a$, the trial is accepted and the oxygen molecule dissociates and fills the two sites, and if there are adsorbed CO molecules on the six NN sites, the oxygen atom reacts randomly with one of CO molecules on its NN sites at once and the produced $CO_2$ desorbs and leaves two vacant sites.

Our simulation is performed on a two-dimensional lattice with $100 \times 100$ sites under a periodic boundary condition. The defects are randomly distributed on the lattice surface with the fractions $p_1$ and $p_2$ of the two types of defects before the simulation begins. We will begin our simulation from an initial state where all sites are vacant, similar to what one does in the experimental process as $y_{\mathrm{CO}}$ is varied back and forth.

On the other hand, in order to change the external control parameter $y_{\mathrm{CO}}$ back and forth in a steplike form, we perform our simulation process by increasing $\Delta t$ Monte Carlo steps (MCS) from $t$ MCS and fixing the parameter $y_{\mathrm{CO}}$. The simulation process will continue after $\Delta y$ is added to $y_{\mathrm{CO}}$, and until $y_{\mathrm{CO}}$ approaches 1.0, $\Delta y$ is then subtracted from $y_{\mathrm{CO}}$ step by step, and $y_{\mathrm{CO}}$ will decline back to the initial value. A cycle simulation process is thus finished.

## III. SIMULATION RESULTS

In our simulation, we exhibit a hysteresis loop and discuss the influences of the inhomogeneities of the surface and external control parameters on the hysteresis phenomena.

Figure 1 shows the macroscopic behavior in the CO catalytic oxidation system when $y_{\mathrm{CO}}$ is varied back and forth (at a constant scan rate $\beta_{\mathrm{CO}} = dy_{\mathrm{CO}}/dt$) and other parameters are kept unchanged. We see from Fig. 1 that for a small fraction $y_{\mathrm{CO}}$, the system is in a state of high reactivity and the O-passivated state is destroyed, in contrast to the classical Ziff-Gulari-Barshad (ZGB) model [17]. As $y_{\mathrm{CO}}$ reaches $\tau_1$, the system transits to a steady state of low reactivity and

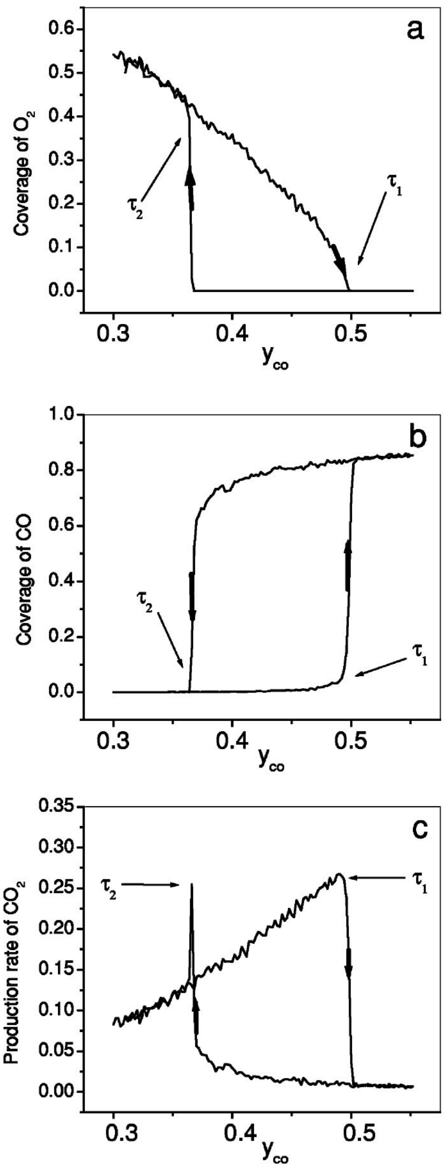

FIG. 1. The simulation results of the hysteresis phenomena about the reactant coverage [(a) and (b)] and production rate of $CO_2$ (c). The simulation parameters are $p_{chem}=0.01$, $p_{ad}=0.7$, $p_{des}=0.1$, $p_{des1}=0.8$, $p_{dse2}=0.05$, $p_a=0.6$, $p_b=0.9$, $\Delta y=0.002$, $\Delta t=10$ MCS, $p_1=0.1$, and $p_2=0.1$.

the surface is mainly covered by adsorbed CO. After the transition, the system remains in the steady state of low reactivity as $y_{CO}$ increases continuously. Furthermore, as $y_{CO}$ begins to reduce, the system continues to remain in the state of the low reactive state even if $y_{CO}$ is less than $\tau_1$. If $y_{CO}$ is lowered to $\tau_2$ ($\tau_2<\tau_1$), the system will exhibit a steep transition from the state of low reactivity to the state of high reactivity. Consequently, a clockwise hysteresis loop forms and the width $\triangle\tau$ of the hysteresis loop is equal to $\tau_1-\tau_2$.

### A. The influence of the defects on the hysteresis behavior and the nucleation and growth of both CO and oxygen islands

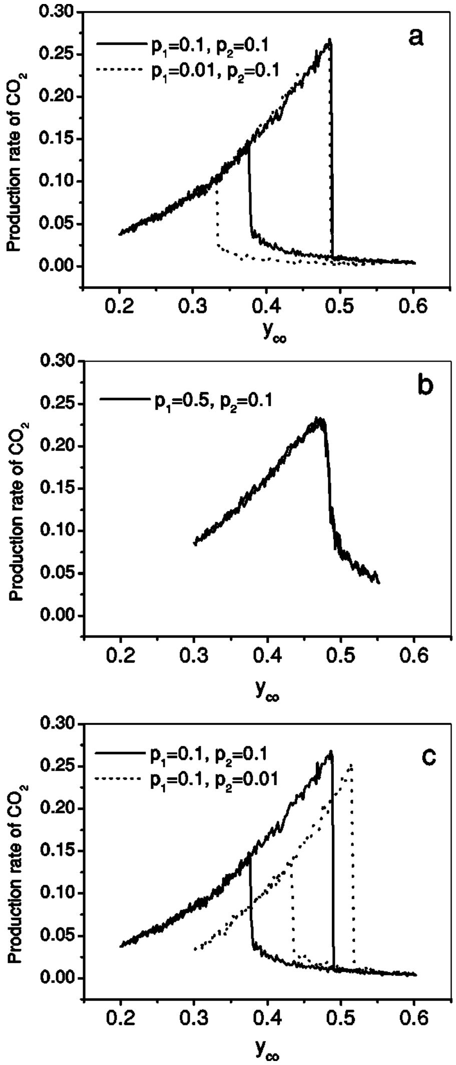

FIG. 2. The influence of the number of the two kinds of defects. In (a) and (b), the width of the hysteresis loop decreases and the loop tends to disappear with the increasing fraction of the type-1 defect. In (c), both $\tau_1$ and $\tau_2$ are affected significantly by the type-2 defect. $\Delta t=100$ MCS and other parameters are the same as those in Fig. 1.

The inhomogeneities of the catalyst surface can have an important effect on the dynamical behavior of the reaction system. In Fig. 2(a), it is shown that the width of the hysteresis loop decreases obviously as the fraction of type-1 defects increases. As the surface is mainly covered by O adatoms, the adsorbed CO almost reacts with the neighbor O adatom and the desorption of CO will not affect the dynamical behavior of the reaction system. Therefore, the change of the fraction of type-1 defects will have little influence on the value of $\tau_1$. However, as the surface is covered by CO and the fraction of type-1 defects increases, the nucleation of adsorbed O atoms becomes easy, and thus $\tau_2$ increases obviously. Most importantly, as shown in Fig. 2(b), the hysteresis loop disappears because the O nucleation and growth of the O island become very easy when the fraction of type-1 defect becomes very large.

Furthermore, the fraction of type-2 defects has a significant influence on the dynamical behavior. As the fraction of type-2 defects decreases, the values of $\tau_1$ and $\tau_2$ increase but the width of the hysteresis loop decreases. When the fraction of type-2 defects becomes small, the sites that cannot be occupied by O atoms become small and the nucleation of adsorbed CO becomes difficult, and thus the transition to the steady state of low reactivity will occur at high values of $y_{\mathrm{CO}}$ [see Fig. 2(c)]. Meanwhile, because the desorption of adsorbed CO on the type-2 defects is less than that on unperturbed sites, $\tau_2$ also increases as the fraction of type-2 defects becomes small.

Now we turn to consider the influences of the two types of defects on the formation of a new state. It has been shown in the previous experimental work by Berdau *et al.* [13] that the "active" inhomogeneities play an important role on creating a new state. Before starting our simulation, we distribute the type-1 and type-2 defects on the surface regularly, i.e., the type-1 defects are distributed on the lattice sites $j = 50$ and the type-2 defects are distributed on the lattice sites $i = 10, 20, \ldots, 100$, $j \neq 50$, where $i$ and $j$ are the positions on the lattice surface. The simulation results are shown in Fig. 3. We see from Fig. 3(a) that there is no apparent effect of the defects on the formation of the CO phase. Actually, from the above simulation results (Fig. 1), the coverage of adsorbed oxygen on the surface is not high before the transition to a state of low reactivity, and thus there are many vacant sites for CO adsorption. Furthermore, the adsorbed CO molecules will diffuse rapidly, leading to a spontaneous nucleation of CO during the transition from the state of high reactivity to the state of low reactivity. Therefore, the type-1 and type-2 defects have little influence on the formation of the CO phase.

However, from Fig. 3(b), we can find that the formation of the O phase on the CO-covered surface is largely affected by the type-1 defects. Before the transition to the state of high reactivity, the surface is almost covered by adsorbed CO, and thus the vacant sites for O adsorption are poor, but the high desorption probability of CO on the type-1 defects will increase the probability of oxygen adsorption. Consequently, the type-1 defects become the centers of O nucleation during the formation of the oxygen phase. This simulation result is in good consistency with the experimental observation [13].

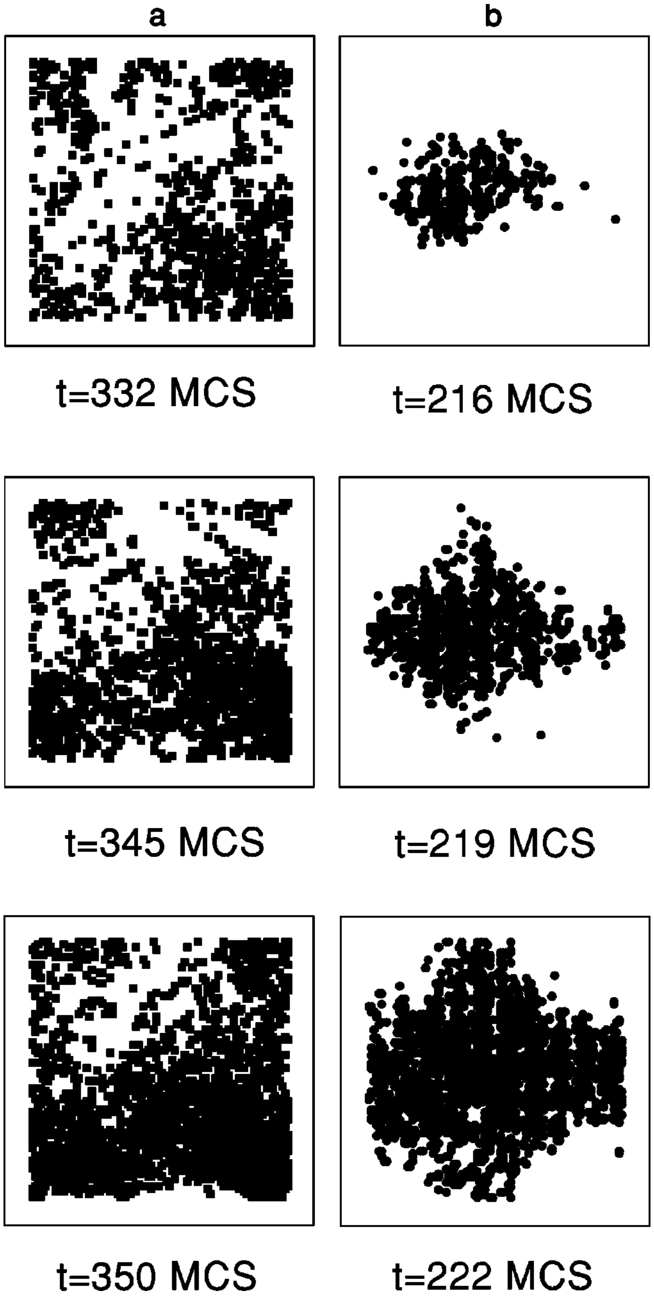

FIG. 3. The formation of a new state. (a) The formation of the CO phase during the transition to a state of low reactivity at $y_{\mathrm{CO}} = 0.500$. (b) The formation of the oxygen phase on the CO-covered surface during the transition to the state of high reactivity at $y_{\mathrm{CO}} = 0.340$. $p_1 = 0.01$, $p_2 = 0.09$, $\Delta t = 500$ MCS, and other parameters are the same as those in Fig. 1.

### B. The dependence of hysteresis behavior on the scan rate $\beta_{\mathrm{CO}}$

According to the experimental results [13], $\Delta\tau$ depends on the scan rate $\beta_{\mathrm{CO}}$ in a way that $\Delta\tau$ decreases with the decrease of $\beta_{\mathrm{CO}}$. Therefore, one may suspect whether the width $\Delta\tau$ approaches zero as the scan rate becomes infinitely low. In the present paper, we can discuss the influences of the scan rate $\beta_{\mathrm{CO}}$ on the width of the hysteresis loop. During the simulation process, we use two ways to change the scan rate. First, we change the simulation time $\Delta t$ for a fixed $\Delta y$ ($\Delta y$ is very small). The second way is to change the $\Delta y$ for a given $\Delta t$. There is no obvious difference for the simulation results under the two different methods.

The simulation results are shown in Fig. 4 for different values of the scan rate $\beta_{\mathrm{CO}}$. From three examples in Figs. 4(a)–4(c), it is exhibited that the width $\Delta\tau$ shrinks as the scan rate becomes small. As $\beta_{\mathrm{CO}}$ becomes small, the steep change at the transition point from low reactivity to high reactivity states will disappear [Figs. 4(b) and 4(c)]. These results are in good consistency with the experimental phenomena [13]. The simulation results for the values of $\tau_1$ and $\tau_2$ are summarized in Fig. 4(d), which clearly exhibits the effect of the scan rate on the width of the hysteresis loop. Figure 4(d) also shows that the width of the hysteresis loop remains a finite constant even if $\beta_{\mathrm{CO}}$ becomes enough small.

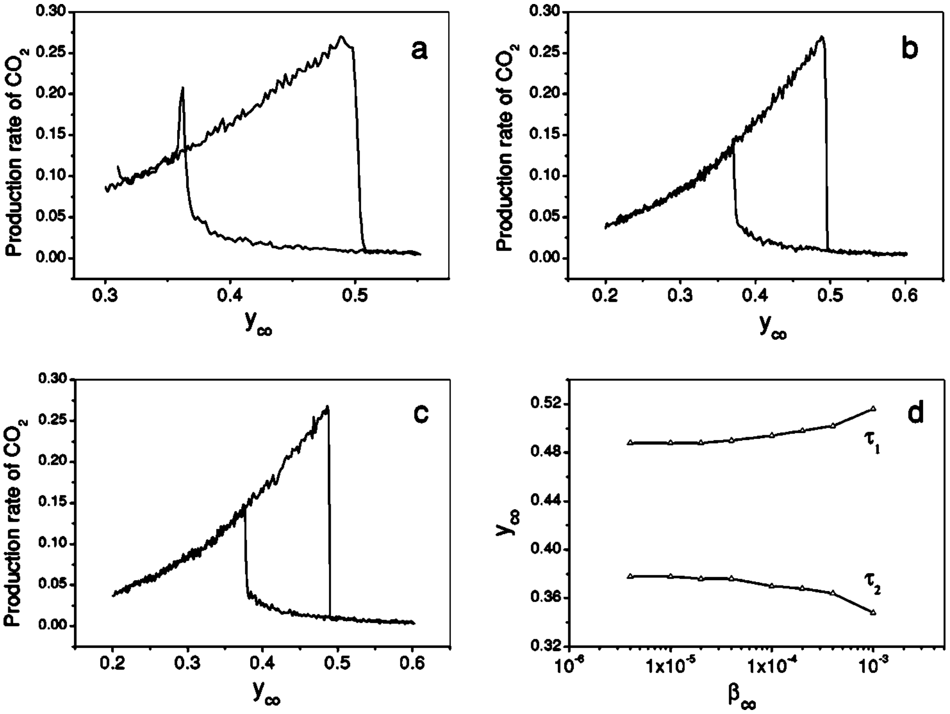

FIG. 4. The effect of the scan rate $\beta_{\rm CO}$ on the width of hysteresis loop. (a), (b), and (c) correspond to $\Delta t=5$, 200, and 500 MCS, respectively. Other parameters are the same as those in Fig. 1.

### C. The important role of the diffusion rate of adsorbed CO

The adsorbed CO diffuses rapidly on the surface and plays a decisive influence on the dynamical behavior of the reaction system [6,28]. We will change the value of $p_{chem}$ to discuss the influence of the diffusion. As illustrated in Fig. 5 through three examples corresponding to $p_{chem}=0.01$, 0.1, and 0.5, we can clearly diagnose that the transition points $\tau_1$ and $\tau_2$ approach each other, and the hysteresis loop shrinks as the diffusion rate decreases. When $p_{chem}=0.5$, i.e., the CO diffusion becomes very slow, it is found that the hysteresis loop disappears. The dependence of the difference between $\tau_1$ and $\tau_2$ on the parameter $p_{chem}$ is summarized in Fig. 5(d), clearly displaying the effect of the diffusion rate. In the following section, we will give a further discussion.

## IV. DISCUSSION

The hysteresis behavior in the CO catalytic oxidation system is exhibited in some related theoretical models which are based on different mechanisms [29,30]. According to the present simulation results, we think that all kinds of proper-

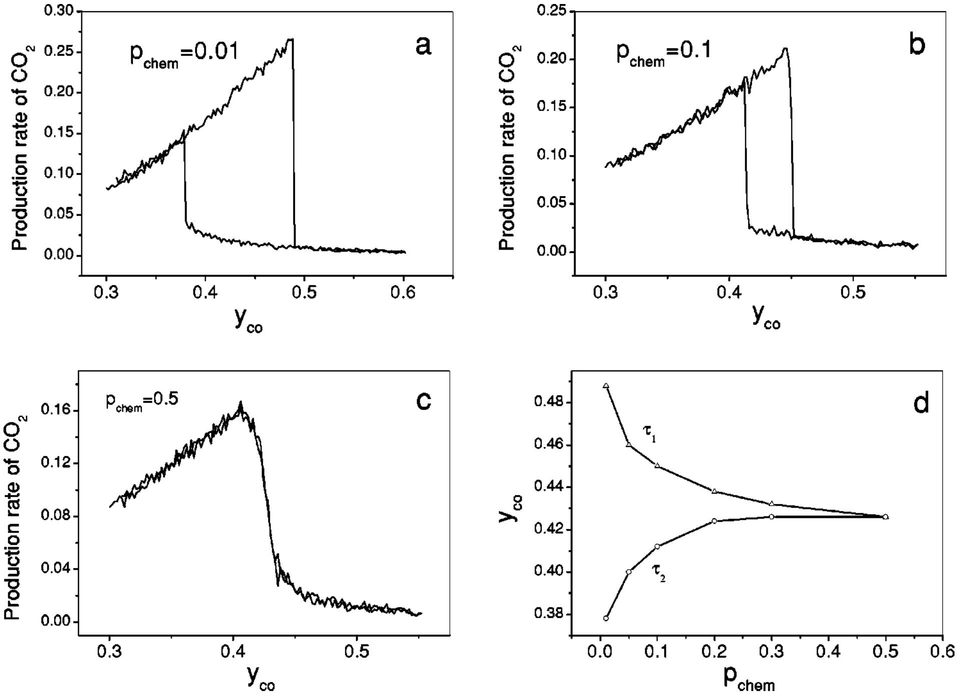

FIG. 5. The influence of the diffusion of adsorbed CO on the hysteresis width. In (a), (b), and (c), $p_{chem}=0.01$, 0.1, and 0.5, respectively. $\Delta t=200$ MCS and other parameters are the same as those in Fig. 1.

ties in hysteresis phenomena are caused by the combination of several factors which are due to the different kinetic behavior of CO and $O_2$ on the catalytic surface.

First, the adsorbed processes of CO and $O_2$ are obviously different. The adsorption of $O_2$ needs more sites on surface than that of CO, and the sticking coefficient of CO is much higher than that of $O_2$. Therefore, if the number of vacant sites on the surface is very small, the adsorption of oxygen is significantly affected. If the surface is mainly covered by oxygen adatoms, CO can adsorb very well and the system shows a steady state of high reactivity. On the contrary, the adsorption of $O_2$ on the CO-covered surface will be very difficult, and the system exhibits a steady state of low reactivity even if the external parameter $y_{\mathrm{CO}}$ is the same as before.

Second, the binding energy of adsorbed CO is much lower than that of the oxygen adatom. Therefore, adsorbed CO can diffuse rapidly on catalytic surface and desorb, but the diffusion and desorption of adsorbed O atom are neglected. As a result, the rapid diffusion of CO makes it easy for adsorbed CO to react with the O adatom and suppresses the nucleation of CO and growth of the CO island on the O-covered surface, and it also suppresses the nucleation of oxygen and growth of the O island on the CO-covered surface. On the other hand, the rapid diffusion of CO on surface makes the distribution of the vacant sites more homogeneous, which makes the adsorption of oxygen more difficult.

## V. CONCLUSION

Using a MC simulation, we have discussed the hysteresis behavior in the catalytic oxidation reaction system in the presence of the inhomogeneities of the catalyst surface. Phenomenologically, in contrast to the classical ZGB model, the O-passivated state is destroyed in our model and a hysteresis loop is exhibited as the external control parameter $y_{\mathrm{CO}}$ is varied back and forth in a steplike form. Moreover, the simulation results show that the defects on the surface have a significant effect on the hysteresis transition points and the formation of the O phase on the CO-covered surface at the transition points.

Furthermore, the present model enables us to study the influences of other parameters on the hysteresis phenomena. First, it is shown that the width of the hysteresis loop shrinks as the scan rate $\beta_{\mathrm{CO}}$ decreases, and there exists a finite loop width of the macroscopic hysteresis even if $\beta_{\mathrm{CO}}$ becomes infinitely small. Second, we examine the effects of the diffusion of adsorbed CO on the hysteresis phenomena, and clearly indicate that the width of the hysteresis loop decreases obviously with the decrease of the diffusion rate, and even hysteresis can be ruled out as the diffusion of adsorbed CO becomes very slow. These simulation results are well consistent with previous relevant experimental results.

## ACKNOWLEDGMENTS

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