Spatiotemporal Patterns During CO Oxidation on Pt(100) at Elevated Pressures

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CO oxidation on Pt(100) was investigated at reactant pressures of 8 Pa using ellipsomicroscopy for surface imaging to monitor spatiotemporal pattern formation on the catalyst surface and mass spectroscopy to monitor the integral reaction rate. Three distinct temperature regions showing nonlinear phenomena were found. The first region, at 540−615 K, is characterized by macroscopic, regular reaction-rate oscillations with spatially uniform, instantaneous transitions between CO and oxygen covered states. Regular, periodic oscillations were not observed on Pt(100) before. The second region, 480−540 K, exhibited continuous dynamic pattern formation with no visible macroscopic kinetic oscillations. Front speeds measured indicated a decrease in oxygen front velocity and an increase in CO front velocity with an increasing CO/O_2 partial pressure ratio. The third region, 390−430 K, is characterized by macroscopic rate oscillations, which continued for several oscillation periods and then ceased with the onset of pattern formation.

Introduction

Oscillatory heterogeneous catalytic reactions have been the focus of intense experimental and theoretical investigation (Slin'ko and Jaeger, 1994). The main goal is to gain a basic understanding of the cause of oscillatory behavior, and therefore to obtain a better insight into fundamental reaction mechanisms. Such studies also have implications for reactor design and process control. Higher reaction rate and selectivity may result if a reaction is carried out in an oscillatory manner (Lagos et al., 1979; Olsson and Schoeoen, 1986). A frequently used model reaction is CO oxidation on platinum group noble metal catalysts. CO oxidation has been studied at pressures less than 0.05 Pa on several well-defined, low-index Pt single crystals, with a view to using simpler systems than supported catalysts (Ertl, 1990, 1993, 1997; Lauterbach et al., 1993; Lauterbach and Rotermund, 1994a,b; Ertl and Rotermund, 1996).

Spatiotemporal pattern formation was observed to exist in rich variety during oscillatory reactions on Pt single crystals at pressures below 0.05 Pa. Patterns have been attributed to the interplay between the various rate processes of adsorption, desorption, reaction, adsorbate diffusion, and adsorbate-induced surface phase transition. It is apparent that pattern formation plays an important role in macroscopic-rate oscillations observed on single crystals. Imaging of surfaces that show such fascinating dynamic phenomena can therefore lead to improvement in our understanding of fundamental mechanisms that are responsible for pattern formation.

Spatially resolved low-energy electron diffraction (LEED) measurements showed, for the first time, that spatial patterns form on Pt surfaces during CO oxidation (Cox et al., 1983). However, LEED has low spatial and time resolution (1 mm and several seconds, respectively), restricting its applicability to patterns on the micron length scale and subsecond time scale. Photon electron emission microscopy (PEEM) has demonstrated much improved time and spatial resolution (Rotermund, 1997a). Both LEED and PEEM are electron-based techniques, and therefore cannot be operated at pressures higher than 0.1 Pa. This creates a pressure gap between the actual operation pressure of most catalytic reactions and experimental investigations at low pressures using electron-based imaging techniques. Recently, ellipsomicroscopy for surface imaging (EMSI) was developed for this purpose

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(Rotermund et al., 1995; Haas et al., 1998). The technique was found to be uniquely sensitive to submonolayer coverages of adsorbates, which qualified it as an ideal tool for studying spatiotemporal patterns at realistic pressures.

In this article, we present the first study of self-sustained pattern formation during CO oxidation on Pt(100) using EMSI. For the first time, regular-rate oscillations are revealed on the Pt(100) surface. This study brings out novel experimental facts about pattern formation at elevated pressures and their relation to global reaction-rate oscillations.

Experimental Setup

The main analysis chamber is a stainless-steel ultrahigh vacuum system pumped to a base pressure below 1×10^{-8} Pa by a 400 L/s ion pump (Riber) and a titanium sublimation pump. This main chamber is equipped with an Auger electron spectrometer (PHI model 10-155 Cylindrical Auger Electron Optics with 32-010 lock-in amplifier and 11-500A Auger system control) and a differentially pumped quadrupole mass spectrometer (Stanford Research Systems RGA100 with electron multiplier), along with an ion gun for argon-ion sputtering. CO oxidation is carried out in a small stainless-steel reactor, which is connected to the main chamber via a straightthrough metal seal valve. Product sampling is performed by leaking the reactor exit stream into the pumped mass-spectrometer head using a variable-conductance valve (Varian) connected to the outlet of the reactor. A transfer rod is used to transfer the sample with sample mount from the reactor to the manipulator in the main chamber and back. The Pt(100) single crystal is mounted vertically in the reaction cell on a combination of linear and rotary feedthrough, which serves as the sample manipulator. Tantalum wires spot-welded directly to the back of the crystal allow resistive heating. Temperature is measured with a type K thermocouple attached directly to the back of the single crystal. Further details of the sample transfer system can be found in another article (Jachimowski and Lauterbach, 1998).

Pressure measurement in the reactor is performed via a capacitance manometer gauge from 10^{-3} Pa to 10 Pa and a convectron gauge from 10^{-2} Pa to atmospheric pressure, both connected to a reducing cross attached to the reactor. Two 2.75 = in. (70-mm) quartz conflat windows are located on opposite sides of the cube to provide access to the laser beam of the EMSI. The reactor is evacuated with a 70-L/s turbo pump (Varian, V70LP) backed by a 250-L/s rotary-vane mechanical pump. The total reactor volume is approximately 0.5 L.

A gas manifold with three mass flow controllers (MKS), two with range 0–20 sccm and one with range 0–50 std. cm^3, is used to regulate the gas flow rates. The reactor pressure is controlled by a MKS 600 series pressure controller, which takes input from the capacitance manometer gauge, and operates a MKS butterfly valve operating above the turbo pump. The system setup allows for independent control of the system pressure, gas composition, and mass flow rates.

The platinum catalyst used for these experiments was a 1-cm-diameter Pt(100) single crystal. The crystal typically underwent argon-ion sputtering for 15 min at an argon partial pressure of 5×10^{-2} Pa, followed by annealing to 1,200 K. Next, the sample temperature was cycled several times between 500 and 900 K in the presence of 5×10^{-3} Pa oxygen. Analysis of the surface chemical composition was frequently performed using Auger electron spectroscopy (AES) to verify the absence of any contaminants on the surface.

The setup for the EMSI and the principle of operation have been described in detail elsewhere (Haas et al., 1996, 1998; Rotermund, 1997; Rotermund, 1997b,c; Dicke et al., 2000). EMSI follows the principles of a nulling ellipsometer. Polarized laser light (wavelength 488 nm) of an argon-ion laser is focused onto the sample surface and the reflected image, after passing through another polarizer and a quarter-wave plate, is recorded with a CCD camera.

Results and Discussion

Parameter space for rate oscillations and pattern formation of Pt(100)

Figure 1 shows the parameter space (defined by the CO/O$_2$ ratio and the corresponding sample temperature at a constant total pressure of 8 Pa) that produces spatiotemporal pattern formation and/or reaction-rate oscillations. Three distinct regions, marked by I, II, and III in Figure 1, exist where nonlinear phenomena could be observed. Details from

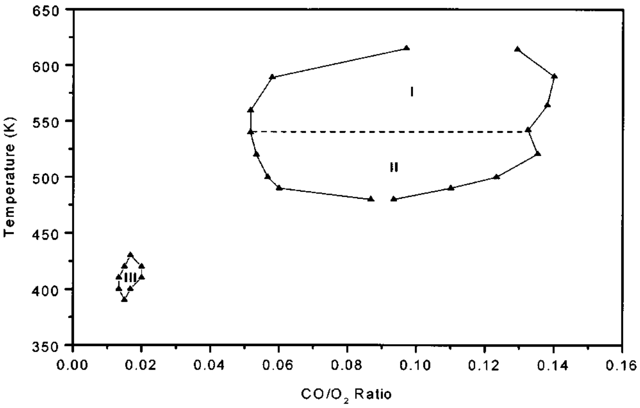

Figure 1. Parameter space of oscillations and/or spatiotemporal phenomena.

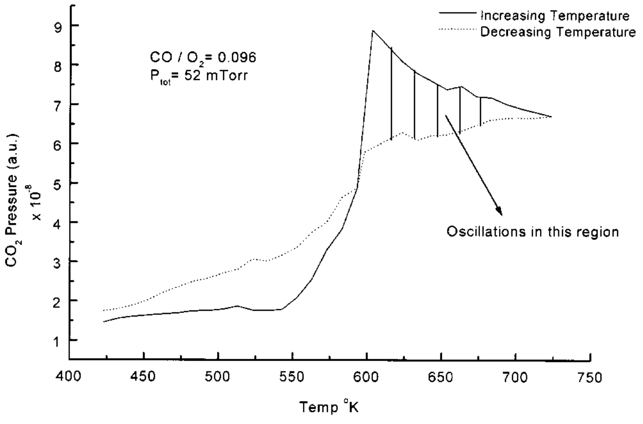

Figure 2. Hysteresis in the reaction rate.

these regions will be discussed in the following paragraphs. All oscillations in the pressure regime were isothermal, as was verified with the thermocouple attached to the sample. It is possible that at these higher pressures, there might exist temperature inhomogenities near the surface; these might not be detected by the thermocouple, which measures the integral temperature at the back of the crystal. However, these effects might be too small to play a significant role in the coverage dynamics on the surface. Prior to the development of the EMSI technique. it was not possible to simultaneously observe both reaction rate and pattern formation in this pressure range. Previous investigations of CO oxidation on Pt(100) report reaction-rate oscillations at pressures below 5×10^{-2} Pa, at temperatures between 460 K and 540 K, and for 2.5 to 10% of CO in oxygen (Cox et al., 1983; Eiswirth et al., 1985; Imbihl et al., 1986). Compared to the studies performed at lower pressures, we observe regular-rate oscillations over a wider temperature range, which is extended to 630 K, although the CO/O_2 ratios in the reactant stream are very similar.

Region I: Reaction-rate oscillations

The first region, (indicated by I in Figure 1) is characterized by regular, large-amplitude oscillations in the production of CO_2. This type of behavior exists at temperatures between ~540 and 630 K, and at CO/O_2 ratios from 0.05 to 0.14. Figure 2 demonstrates the variation in the reaction rate during a temperature ramp for a constant CO/O_2 ratio of 0.096 for regime I. A hysteresis behavior is observed, that is, the reaction rate follows a different path when the temperature is increased compared to when it is decreased. When measuring the increasing temperature curve, the system starts from a completely CO-covered state, which inhibits reaction by preventing dissociative adsorption of oxygen (CO-poisoned state). Higher temperatures promote desorption of CO, which shows a desorption maximum around 520 K (Lauterbach and Rotermund, 1994a). Around this temperature, the equilibrium coverage of CO becomes low enough for oxygen to adsorb, and the reaction rate starts to increase. The maximum reaction rate is reached around 600 K, and then decreases again with further temperature increase. The reaction-rate maximum coincides with the temperature for the onset of desorption of oxygen from the surface (Lauterbach and Rotermund, 1994a) and decreases with increasing temperature, mainly due to the overall decrease in oxygen coverage. When decreasing the temperature from the high reactive state, the surface starts from a high reactive, mainly oxygen-covered state with low total coverage. In this state, the rate-limiting steps are predominantly the adsorption/desorption of the reactants. On this path, no distinct maximum in the reaction rate is observed, and the reaction-rate curve crosses the increasing T branch at temperatures around the reaction-rate maximum. This could be due to asymmetric inhibition of the oxygen adsorption, where CO still inhibits the adsorption of oxygen on the increasing branch, while oxygen can readily adsorb in the decreasing branch due to the lower overall coverages. Hysteresis behavior has previously been reported for several single-crystal surfaces as the CO partial pressure was changed for constant temperature and oxygen partial pressure (Berdau et al., 1997; Kokodziejczyk et al., 1997), and typically the dependence of the reaction rate upon the CO concentration is observed to pass through a maximum.

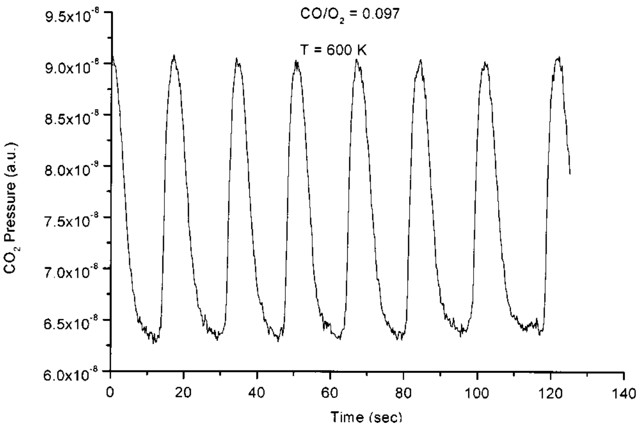

Figure 3. Regular-rate oscillations in Region I at 600 K, $CO/O_2 = 0.097$.

Region I is characterized by very regular-rate oscillations (Figure 3). The period and the shape of the oscillations remain constant even over hours of observation. EMSI reveals that in this region, transitions from the CO to the oxygen-covered state and vice versa for periodic oscillations are spatially uniform and occur instantaneously. Essentially, the surface flip-flops between states within one video frame (1/30 s). Periodic oscillations of this type could not be observed under low-pressure conditions (Lauterbach and Rotermund, 1994a). Relaxation oscillations were also observed in regime I (Figure 4). These oscillations, however, are now accompanied by distinct pattern formation. Transitions from the high reactive to the low reactive state are very fast (within one video frame), and are still spatially uniform with no visible pattern formation. However, the reverse transition resulting in the return to the high reactive state now occurs via oxygen fronts forming on the predominantly CO-covered surface and quickly spreading into the CO-covered areas. Between transitions, the

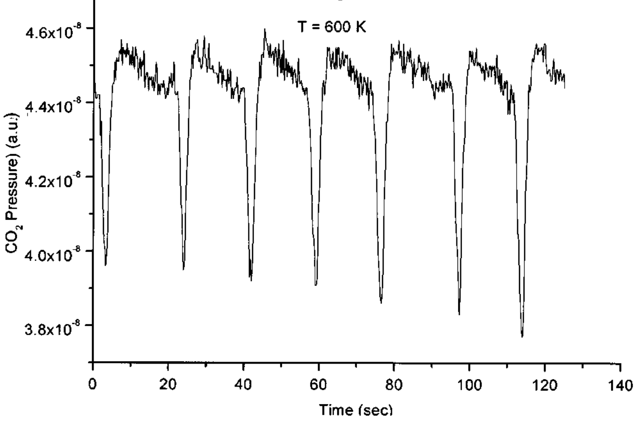

Figure 4. Relaxation oscillations in Region I at 600 K, $CO/O_2 = 0.062$.

system now remains longer in the high reactive state, leading to the relaxation oscillations. Again, such oscillations could never be observed under low-pressure conditions on the Pt(100) surface (Lauterbach and Rotermund, 1994a).

An increase in temperature typically leads to a decrease in the period of oscillations (see, for example, Figures 4 and 5), as has been observed for the CO oxidation for other single-crystal surfaces. The EMSI again reveals spatially uniform transitions from the CO-covered state to the oxygen-covered state and back. Increasing the CO/O_2 ratio to 0.14 (Figure 6) leads to an increase in the period of oscillations. This is also expected, as the surface now stays in the CO-poisoned state for a longer time owing to higher adsorption rates of CO compared to the oxygen adsorption rates.

During oscillations in regime I, the entire single crystal is coupled efficiently through the gas phase. Every part of the catalyst surface effectively sees the same number of molecules of reactants. This results in synchronization of various areas, leading to well-defined reaction-rate oscillations. This long-range coupling mechanism is necessary for global-rate oscillations, and has also been reported for Pt(110) and polycrystalline platinum foils at lower pressures (Falcke and Engel, 1994; von Oertzen et al., 2000; Lauterbach and Rotermund, 1994b).

Region II: Pattern formation in the absence of oscillations

Pattern formation without the presence of large-amplitude observable macroscopic-rate oscillations characterizes the second regime (II). This phenomenon occurs between temperatures of 480 and 550 K. The presence of self-sustained dynamic phenomena on the catalyst surface pattern formation without any observable oscillations in the reaction rate demonstrates the need for coupling mass spectrometry with imaging, as this dynamic behavior would remain undiscovered in the absence of imaging. Figure 7 demonstrates one of the several types of patterns observed in this region. In the first six images, CO islands grow via slow-moving CO fronts with velocities of ~ 25 micron/s. The last four images show a higher velocity oxygen front (~ 250 micron/s) moving across the surface and transforming it back into the predominantly oxygen-covered state. It was observed that oxygen fronts mainly originate at either surface defects or the edge of the single crystal, which has a higher defect density.

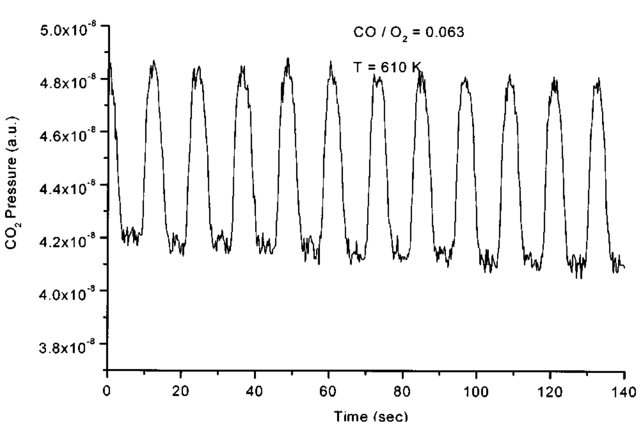

Figure 5. Regular-rate oscillations in Region I at 610 K, CO/O_2 = 0.063.

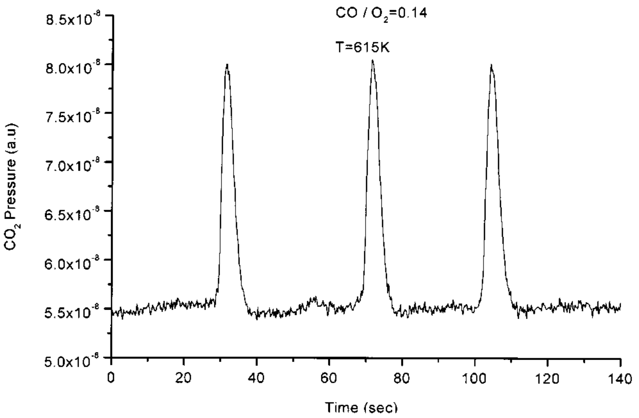

Figure 6. Regular-rate oscillations in Region I at 615 K, CO/O_2 = 0.14.

The absence of measurable reaction-rate oscillations can be explained as a result of the nature of the pattern formation. For global reaction-rate oscillations to occur, the surface must undergo transitions in which the majority of the surface becomes primarily occupied by one adsorbate species, followed by a reverse transition during which the other adsorbate occupies the surface. When spatiotemporal patterns exist on the surface, such as in regime II, there are many areas present in either the high or low reactive state. This eliminates the possibility of macroscopic, large-amplitude-rate oscillations. The coupling of different active areas on the catalyst surface in this region is governed mainly by local surface diffusion. The long-range coupling mechanism, as proposed for regimes I, is not very effective in regime II.

However, it might also be possible that small-amplitude oscillations or fluctuations cannot be detected due to residence-time effects in the mass spectrometer and the limited temporal resolution of the mass spectrometer of ~ 2 data points/s. For this purpose, the average brightness of the EMSI images was measured as a function of time. Figure 8 shows the brightness levels for oscillations from regime I and for pattern formation in regime II. While for regime I (curve A), the macroscopic-rate oscillations are clearly reflected in the EMSI brightness, the data from regime II (curve B) show some fluctuations in the reaction rate, which could be interpreted as small, irregular oscillations. For lower pressures, irregular oscillations for this reaction on Pt(100) have been observed, albeit with much larger amplitude (Lauterbach and Rotermund, 1994a; Imbihl et al., 1986).

Front velocities in regime II were also determined using the recorded EMSI images. Data showing corresponding velocities for oxygen and CO in regime II are shown for different temperatures (see Figures 9 and 10). The velocity of oxygen fronts decreases and that of CO fronts increases with an increase in the CO/O_2 ratio. At higher CO/O_2 ratios, CO adsorption dominates and leads to higher CO coverages on the surface. This restricts adsorption of oxygen, leading to a decrease in the front velocity of oxygen. The same effect can explain the increase in oxygen front velocity with decreasing

CO/O_2 ratio. Both front types show an increase in the front velocity with increasing catalyst temperature, which is expected, since the diffusion of adsorbates, in particular CO, on the surface increases with temperature.

Oxygen front velocities in this study were 4 to 5 times higher than those measured during lower-pressure studies (Lauterbach and Rotermund, 1994a), while CO front velocities were only up to two times faster. At 8 Pa, front velocities on Pt(100) reached values up to 500 micron/s for oxygen fronts and up to 40 micron/s for CO fronts, coupling large areas of the single-crystal surface very efficiently. At higher pressures, higher fractional coverages are expected on the surface, leading to higher reaction rates between CO and adjoining oxygen. Since each CO_2 molecule formed is a result of two sites being vacated by oxygen, as against only one by CO molecules, the oxygen fronts travel faster compared to CO.

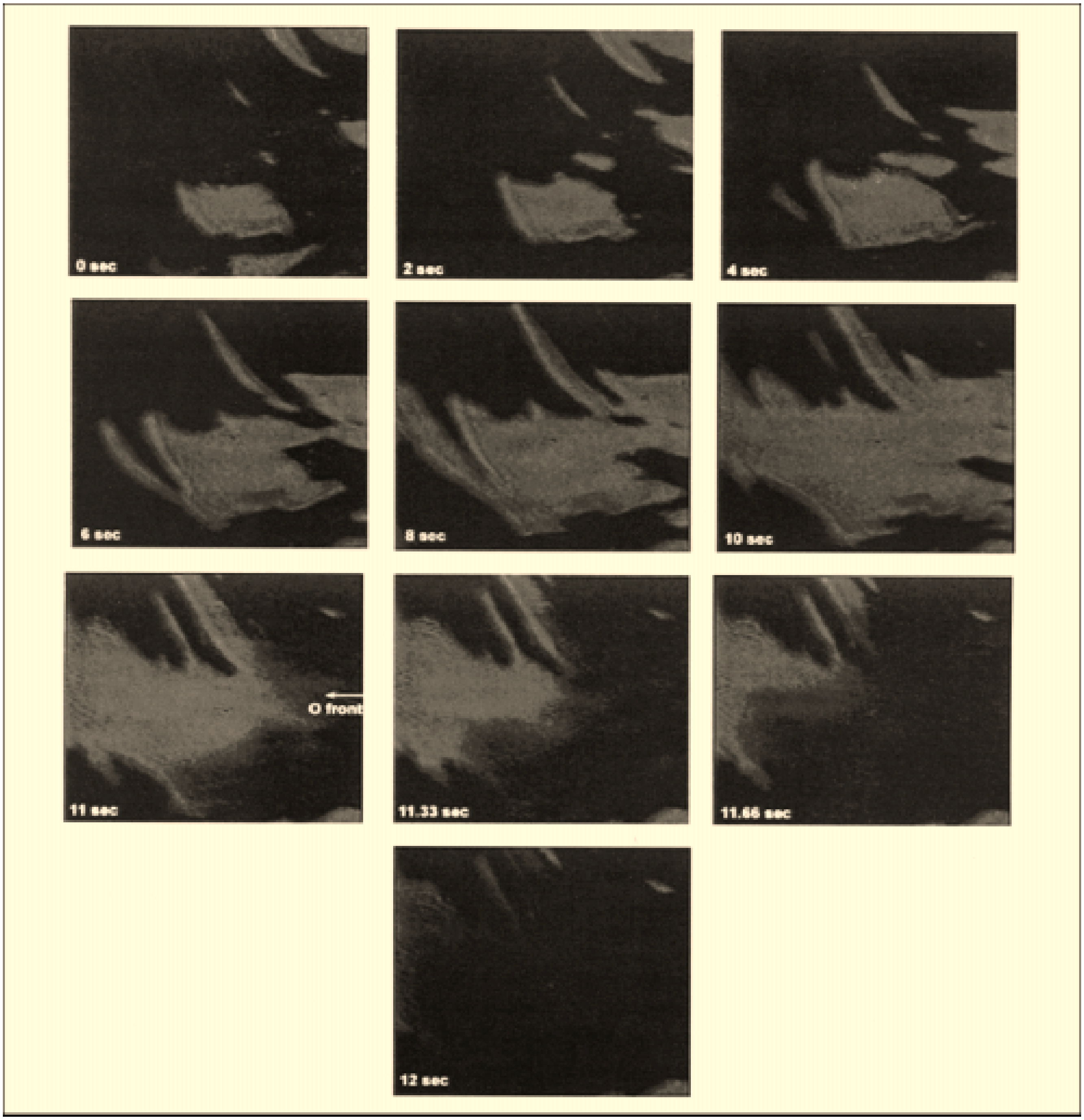

Figure 7. Sequence of images (at intervals of 2 s) of the Pt(100) surface taken with the EMSI in region II at 520 K, $CO/O_2 = 0.09$.

Oxygen-covered areas appear darker, while CO-covered areas are lighter; the images size is ~1 mm × 1.25 mm.

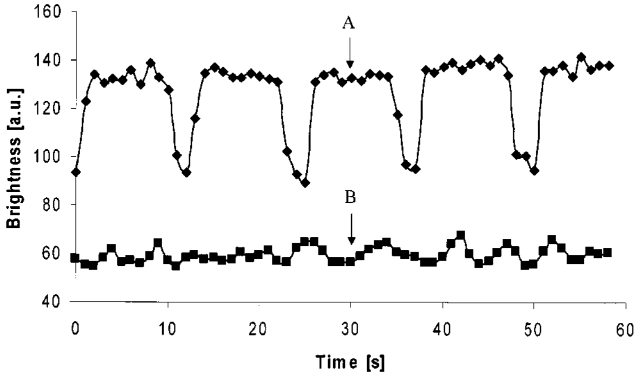

Figure 8. Averaged brightness of the EMSI image during (A) oscillating reaction rate, with the surface alternating between CO-covered and oxygen-covered states, and (B) random pattern formation when the rate oscillations are not detected.

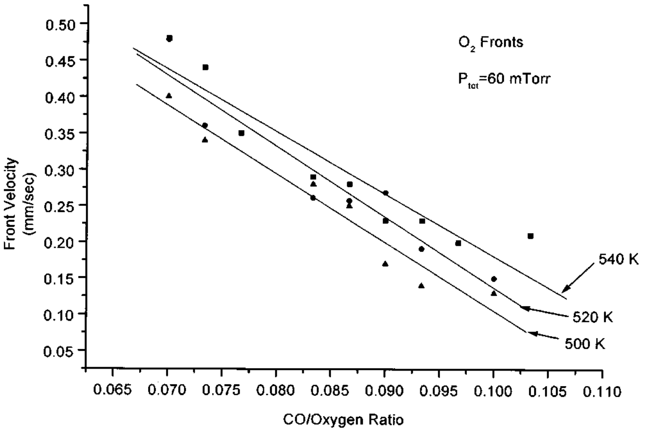

Figure 9. Oxygen front speeds measured in Region II.

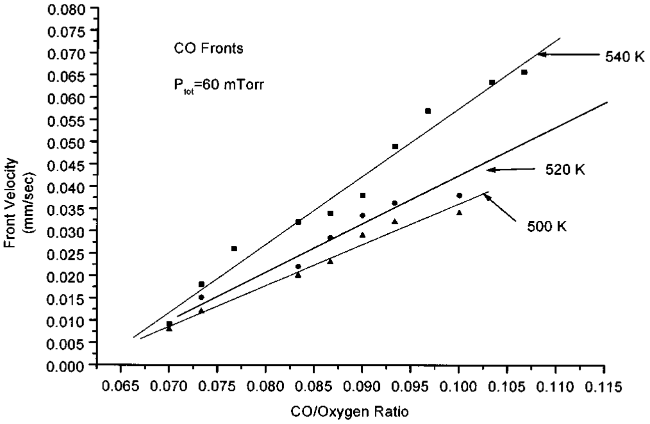

Figure 10. CO front speeds measured in Region II.

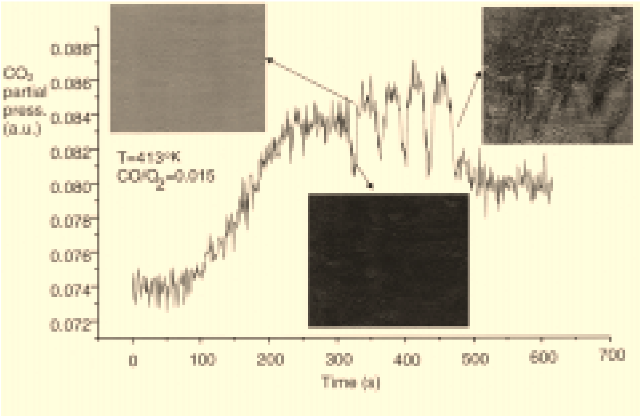

Figure 11. Rate oscillations and EMSI images demonstrating behavior in Region III.

$T = 410$ K, $CO/O_2 = 0.015$; image size is ~1 mm × 1.25 mm.

Region III—Unsustained rate oscillations

Spatiotemporal pattern formation was also observed in region III between 390 and 430 K, at considerably lower temperatures than the two other regions of pattern formation. Region III is not connected in the parameter space with regions I and II, that is, for parameters between combined regions I and II and region III, no nonlinear phenomena could be observed. Periodic-rate oscillations, similar to those seen in region I, were found to exist. However, the rate oscillations were not self-sustained indefinitely and typically ceased within several minutes. Figure 11 shows the variation of the reaction rate with the corresponding EMSI images, which show images at specific points during the oscillatory cycle. After establishing the reaction parameters, the system undergoes a rather slow transition to a high reactive state. After remaining in this state for approximately 30 s, the system begins to oscillate. The reaction rate indicates fast oscillations characterized by a transition to the low reactive state, followed by an immediate reverse transition back to the high reactive state. The EMSI data reveal that the forward transition is again spatially uniform and with no observable pattern formation. The reverse transition initially occurs as a homogeneous change; however, with time it begins to show pattern formation, which becomes more prominent with each oscillatory cycle. Patterns develop with oxygen fronts growing mainly from surface defects and overtaking the predominantly CO-covered surface. The fronts now grow in elliptical shapes, indicating that the front velocities in this temperature regime have some relationship with the orientation of the crystal. Such orientation effects could not be observed for regimes I and II. Eventually, reaction-rate oscillations cease when a forward transition to the CO-covered state occurs and oxygen fronts are unable to reclaim the surface to produce the reverse transition, leaving the surface in a low reactive, CO-covered state, and shows nonlinear phenomena at much lower temperatures than previous studies (Lauterbach and Rotermund, 1994a).

Summary

EMSI coupled with mass spectrometry is an effective experimental combination in accessing concentration pattern information and reaction-rate data at arbitrary pressures. This was demonstrated for the case of isothermal CO oxidation of Pt(100), where nonlinear reaction behavior at intermediate pressures was studied. Three distinct regions of rate oscillations and/or spatiotemporal pattern formation were found to exist. The first region was characterized by macroscopic, regular reaction-rate oscillations with homogeneous instantaneous transitions on the Pt(100) surface, which we attribute to an effective global coupling through the gas phase. Only irregular oscillations have been observed on Pt(100) at lower pressures before, leading to the conclusion that the global coupling through the gas phase is enhanced at higher pressures. The second region exhibited continuous dynamic pattern formation with no visible kinetic oscillations. The third region was characterized by macroscopic-rate oscillations, which continue for several periods and then cease with the onset of pattern formation in the reverse transition.

Compared to previous studies of CO oxidation on Pt(100), performed at pressures two to four orders of magnitude lower, novel behavior has been observed, demonstrating that total reactant pressure alone can have a pronounced influence on the spatiotemporal pattern formation on single-crystal surfaces.

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