Formation and Dynamics of a Hot Zone in Radial Flow Reactor

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An infrared camera was used to study the formation and dynamics of hot zones normal to the flow direction on the surface of a radial-flow reactor, placed horizontally, inside a vessel. The test reaction—the atmospheric oxidation of carbon monoxide—exhibited steady-state multiplicity. Slow cooling of the vessel caused a shift from a fully-ignited state to one with a hot zone in the center of the reactor bounded by cold zones at both reactor ends. The hot zone was separated by sharp temperature fronts from the adjacent cold zones. It shrank upon further cooling of the vessel and eventually extinguished. In certain cases, two stable states existed for the same operating conditions with qualitatively different hot zones and reaction rates. A slow back-and-forth motion of the temperature fronts (breathing) caused complex oscillations in the overall reaction rate. The dynamic features of the two fronts were different in some cases due to nonuniform catalytic activity and inefficient thermal dispersion. The amplitude and frequency of the temperature breathing motion increased upon cooling of the vessel containing the reactor. Experiments suggest that transversal hot zones are likely to form in packed-bed reactors close to the extinction temperature.

Introduction

It is well established that transversal hot zones, with a temperature much higher than that of the adjacent zones, may exist in packed-bed reactors. Reports of the existence of hot zones in industrial reactors were reported by Jaffe (1976) and Barkelew and Gambhir (1984). Boreskov et al. (1981) presented a map of the temperature distribution in a 60-cm-dia. reactor that showed several hot zones existed at the bottom of the reactor. Moreover, hot zones are known to appear and disappear periodically (flickering) on a catalytic gauze during the synthesis of hydrogen cyanide. The presence of hot regions may have a deleterious impact on the yield of the desired product in a packed-bed reactor and may deactivate the catalyst. Moreover, they may lead to severe safety problems by either initiating highly exothermic reactions (the rate of which is negligible under normal operating conditions), or by decreasing the strength of the reactor wall if the hot zone is next to it.

Several potential causes have been proposed to explain the occurrence of hot zones in packed-bed reactors. The first is nonuniformity in the packing of the reactor or the activity of the catalyst. Boreskov et al. (1981) reported hot zone formation due to nonuniform packing of the catalyst. The second potential cause is a nonuniform flow field generated by the interaction between the chemical reaction and the change in the physical properties of the fluid (such as viscosity and density). Matros and Chumakova (1980) and Lee et al. (1986) have shown that reaction-induced change in the fluid properties may lead to different steady-state temperatures and velocities in different tubes in a multitube reactor. Colin and Balakotaiah (1994), Nguyen and Balakotaiah (1994), and Benneker et al. (1998) showed that this type of interaction may generate flow maldistributions and local hot zones in packed-bed reactors. Using a psuedo-homogeneous model, Balakotaiah et al. (1999) predicted that transversal hot zones may form in a packed-bed reactor when the radial dispersion of mass exceeded that of the radial heat dispersion. This condition is analogous to the requirement that the length scale of the inhibitor will be larger than that of the autocatalytic variable in order to obtain a nonuniform state in a diffusion-reaction system (Field and Burger, 1985; Horvath et al., 1993). Yakhnin and Menzinger (2001) pointed out that this condition usually is not satisfied in packed-bed reactors. At pre-

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sent, no criteria or analysis exist to predict the operating conditions, reactions, and reactor configurations that generate hot zones for a realistic set of parameters. The inherent nonuniform activity of commercial catalyst pellets and nonuniform packing of the catalyst in commercial reactors are likely to create even more complex temperature patterns than those in a uniform-activity bed.

Knowledge and understanding of the rate processes leading to the evolution of transversal hot zones is needed for development of criteria predicting their existence and impact, and operating procedures that avoid their formation. There is a definite need for experimental data on the conditions leading to formation of these hot zones and their dynamic features to enhance the ability to predict conditions, reactions, and reactor configurations likely to generate them. Recently, we initiated an experimental study of hot zone formation normal to the flow direction in packed-bed reactors. The experiments involved temporal monitoring of the surface temperature of a thin radial flow reactor by an IR camera during the atmospheric oxidation of carbon monoxide. Heat losses and wall conduction tend to have a strong impact on the evolution, stability, and motion of hot zones in laboratory reactors. Our reactor was made of reticulated alumina cylinder on which Pd was impregnated. The absence of reactor walls circumvented the impact of wall heat conduction that distorts the surface temperature of laboratory reactors, and dampens the amplitude and frequency of the surface temperature oscillations.

Experimental Setup and Procedure

Carbon monoxide was oxidized under atmospheric conditions in a radial flow reactor at vessel temperatures in the range of 35°C–250°C. The reactor was a hollow cylinder (25 mm i.d., 32 mm o.d., 102 mm long) of reticulated alumina (SALI; 80% alumina, 20% silica, 84% open porosity; Zircar) on which 0.95% wt. palladium was impregnated from a solution of 10 wt. % palladium(II) in 10 wt. % nitric acid (Aldrich). After impregnation, the support was calcined at 600°C in air for 1 h and then reduced at 200°C for 8 h. The 10.6 g hollow, cylindrical catalyst had a dark brown coloration, a BET surface area of 45 m^2/g, an average pore size of 150°A, and a density of 480 kg/m^3. The reticulated alumina hollow cylinder was used as the catalyst bed, because it has no walls: the conduction through walls distorts temperature patterns. Two nonporous alumina discs were cemented on both ends of the hollow cylinder to ensure radial flow. The reactor was kept in a horizontal position to minimize the influence of nonuniform heat transfer by natural convection. It was supported on the feed tube that passed through the two nonporous alumina discs at the ends of the reactor. The feed was introduced through a porous steel tube in the center of the reactor to ensure uniform flow distribution. The exterior catalyst surface temperature was monitored through a sapphire window by an IR camera.

The reactor was housed in a rectangular stainless steel vessel (11.7 cm x 11.7 cm and 20.3 cm long) having an infrared-transparent sapphire window (Rayotek) on one side (see Figure 1). This vessel was electrically heated from the outside, and was thermally insulated. The vessel wall temperature was measured by thermocouples and regulated by a temperature controller (PID, Omega CN2041). The composition of the vessel effluent (CO) was measured by an infrared gas analyzer (AR-411; Anarad) at a sampling frequency of 0.1 Hz.

The exterior reactor surface temperature was measured by an infrared camera (Amber Radiance PM) having a 256×256 indium-antimonide detector array, sensitive to 3–5 μm radiation. The field of view of the 50 mm lens was 11° x 11° and the spatial resolution was 0.4 mm^2. A pair of gold-plated mirrors identical to each other were placed behind the catalyst to enable a simultaneous front and back vision of the reactor. The mirrors reflected the infrared radiation and enabled monitoring of the radiation from the back of the reactor. The spatial temperature was recorded on a computer using Imagedesk II (software; Raytheon Amber). Each image consisted of three sections that are shown in Figure 1: the front of the bed (b), and the two reflected side images (a, c). The images of the three sections were processed to show the temporal surface temperature as a function of the angular position and length (Figure 1).

The feed gases consisted of purified grade nitrogen (purity 99.998%), extra dry grade oxygen (purity 99.6%), and carbon monoxide (Aeriform). The carbon monoxide was passed through a carbonyl trap of a molecular sieve adsorbent (5A zeolite; Linde) kept at 240°C. The gas-flow rates were regulated by thermal mass-flow controllers (FC-280, FC-261, and FC-260; Tylan; accuracy $\pm 1\%$). The feed gases were mixed in a bed of glass beads, purified, and dried by activated charcoal purifiers (Linde) before entering both sides of the feed tube. The effluent exited through four outlet ports (4.7 mm i.d.) at the two square ends of the steel vessel.

During this experiment, the vessel temperature was slowly increased until the whole reactor was ignited and its surface attained a uniform high temperature. When the reactor was fully ignited, the vessel temperature was stepwise decreased until a uniform extinguished state was obtained. At each vessel temperature, after a stationary state was reached (1.5 to 2 h), infrared images were recorded. A typical run took 24 h, with 75% of the total data taken close to the extinction point, as states with hot zones existed only in that region. Infrared images were recorded at the rate of 1 per min. to capture effectively the slow hot zone motion. The feed in most experiments contained 6 vol. % CO, 70 vol. % O_2 and 24 vol. % N_2.

In earlier work (Marwaha et al., 2001) we found that an interaction (global coupling) existed between the effluent in the vessel and catalyst surface. This interaction stabilized temperature patterns and complex motions that did not exist in its absence. This conclusion was verified by carrying out experiments where vacuum was used to rapidly evacuate the effluent from the vessel. In order to reduce this interaction we surrounded the reactor in all the experiments reported here with a quartz (infrared transparent) sleeve, with a 2–3 mm gap between the catalyst surface and quartz tube. The volume of the effluent in the sleeve of 21 cm^3 was such that the residence time of the gas in it was less than 1 s, so that its impact on the very slow motion of the hot zone must have been small. The temperature patterns obtained under the same total flow rate (1,600 cc/min) and feed composition (6 vol. % CO, 70 vol. % O_2, 24 vol. % N_2) show that the size of the hot zone was smaller when the quartz sleeve was not used

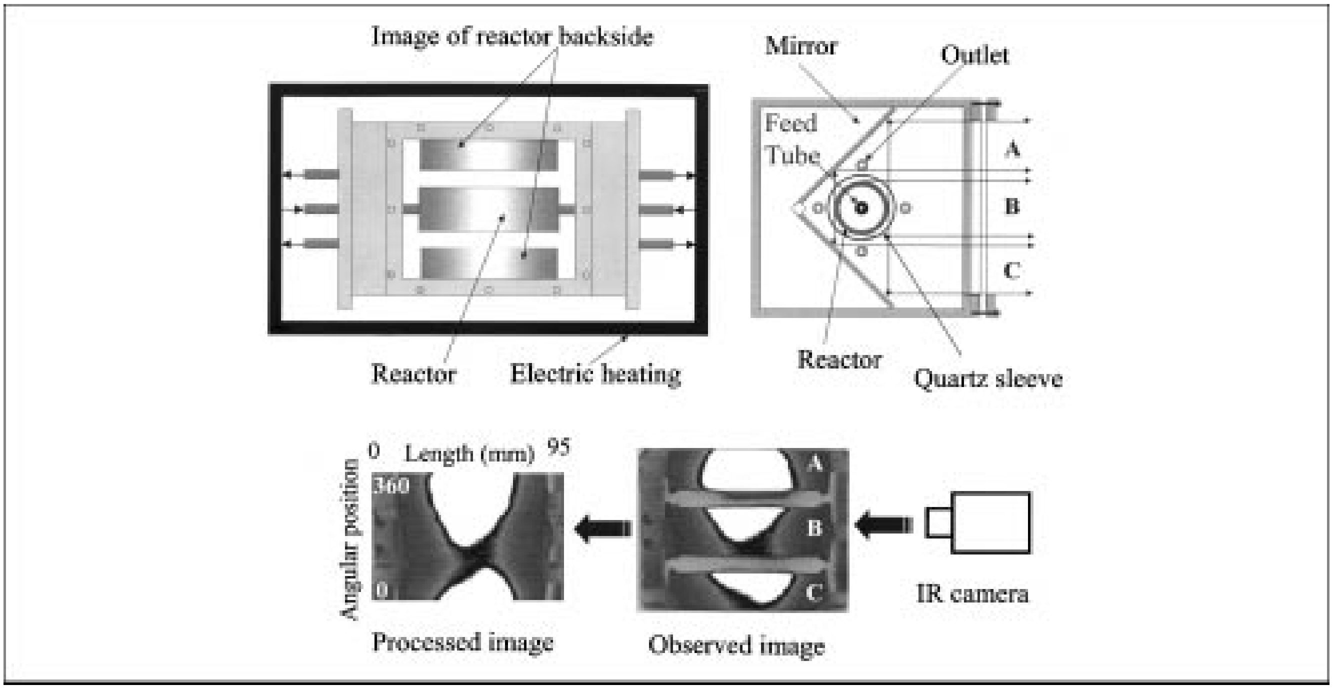

Figure 1. Top: front view of the vessel housing the reactor and side view of the two mirrors enabling simultaneous monitoring of the front and back of the reactor; bottom: observed reactor image recorded by the IR camera and the processed image of the instantaneous temperature as function of reactor angular position and length.

(Figure 2a), than when it was used (Figure 2b). In addition, the amplitude and frequency of the back-and-forth motion of the sharp temperature fronts separating the hot and cold regions decreased when the quartz sleeve was used. The temperature of the vessel housing the reactor is denoted as T_V in all the figures.

Experimental Results

Both extinguished and fully ignited states (bi-stability) were observed during the oxidation of a mixture of 6 vol. % CO, 70 vol. % O_2 and 24 vol. % N_2 over a bounded range of temperatures (Figure 3). We define a state to be *fully ignited*

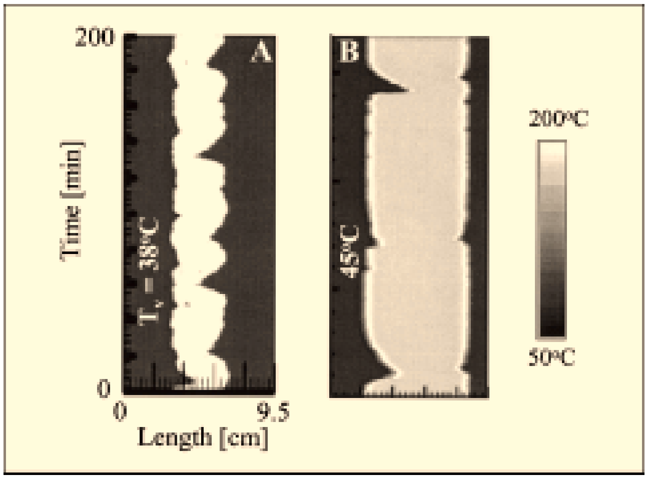

Figure 2. Spatiotemporal temperature patterns obtained with the same reactants flow rate and operating conditions with (B) and without (A) a quartz sleeve surrounding the reactor.

(T_V = vessel temperature).

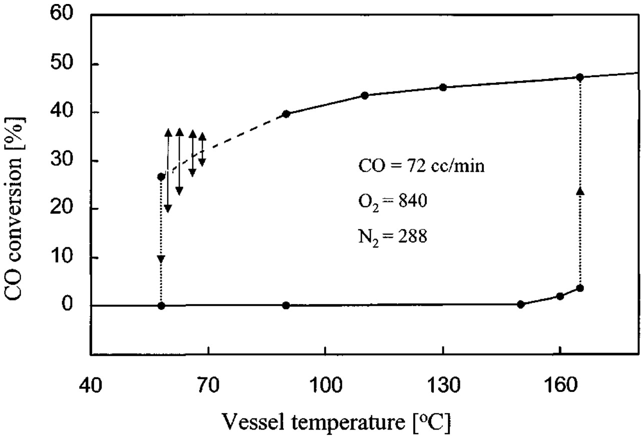

Figure 3. Bifurcation diagram of exit CO conversion as a function of vessel temperature for a feed of 6, 70, and 24 vol. % of CO, O_2 and N_2, respectively, at a flow rate of 1,200 cc/min.

Double-headed arrows describe the oscillations amplitude of the effluent CO concentration of states with a hot zone and dual-front motion.

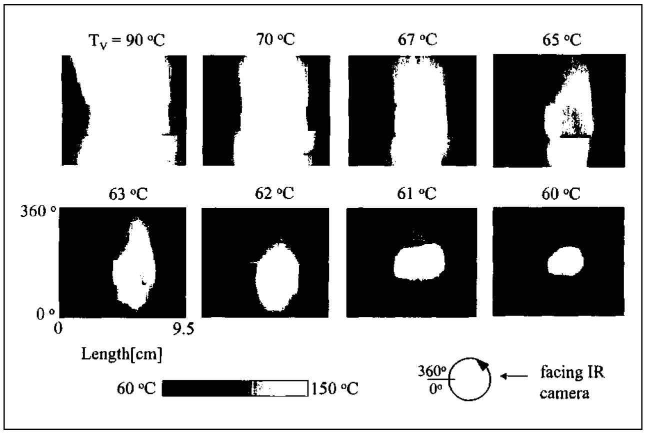

Figure 4. Evolution of the hot zone following a slow gradual decrease of the vessel temperature (T_V).

Same operating conditions as those in Figure 3.

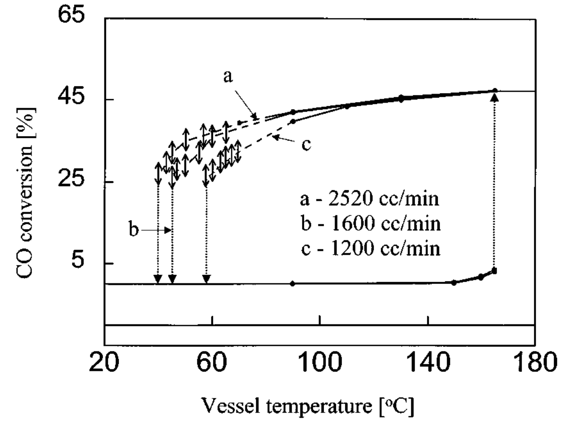

Figure 5. Dependence of CO conversion on vessel temperature for three different feed flow rates.

Feed contains 6 vol. % CO, 70 vol. % O_2 and 24 vol. % N_2. The double-headed arrows denote the amplitude of states with hot zone and dual-front motion.

when the whole reactor surface is at a high temperature, and *extinguished* when the entire reactor surface is at a low temperature. The effluent CO concentration of the ignited states was insensitive to the vessel temperature at temperatures close to and exceeding the ignition point (165°C). As the vessel was cooled, the temperature of the reactor became nonuniform, as illustrated by the map of the instantaneous reactor surface temperature (Figure 4). Initially, two cold regions formed next to the reactor supports (Figure 4, $T_v =$ 90°C) with a sharp temperature front separating the cold and hot regions. The temperature front width, defined as the ratio of the temperature difference between the two zones divided by the maximum temperature gradient, was about 4–7 mm. The hot region shrank following further cooling of the vessel (Figure 4, 70°C–63°C), and was eventually completely surrounded by the cold region (Figure 4, 63°C). Further cooling of the vessel eventually led to hot zone extinction. These nonuniform states are denoted by dashed lines in all the bifurcation diagrams. As the size of the hot region shrank upon cooling of the vessel, the temperature fronts surrounding the hot region started a back-and-forth (breathing) motion and the corresponding effluent CO concentration oscillated. The amplitude and frequency of the back-and-forth motion of the sharp temperature fronts separating the hot and cold regions increased upon cooling of the vessel. In most of the experiments both temperature fronts exhibited breathing motion. However, in some experiments reported later only one temperature front moved back-and-forth. In the figures the states with oscillating CO concentrations are denoted by arrows. A double-headed arrow denotes a state in which both fronts moved back-and-forth. A single-headed arrow denotes a state in which only one front moved back and forth.

The nonuniform states were always observed at vessel temperatures close to the extinction of the reactor. In most cases the transition from the fully ignited states to the nonuniform states was supercritical, as evidenced by a smooth change in the CO conversion (Figure 3). It is well known that the flow rate can affect the extinction temperature of the branch of uniformly ignited states. Experiments carried out with identical feed composition (6 vol. % CO, 70 vol. % O_2 and 24 vol. % N_2) revealed that increasing the total flow rate decreased the vessel temperature causing extinction, but had practically no impact on the ignition temperatures (Figure 5). These experiments indicate that changes in the flow rate may cause a transition between the uniform and nonuniform temperature states. For all the flow rates shown in Figure 5, both temperature fronts moved back-and-forth on the surface when the system attained an oscillatory nonuniform state.

Changes in the CO concentration (at a fixed CO/O_2 ratio) affected the qualitative features of the bifurcation diagrams and of the dynamic features of the temperature fronts. These changes are apparent by comparing the bifurcation diagram (Figure 3) with Figure 6, obtained for a feed of 2.5, 28.8 and 68.7 vol. % of CO, O_2 and N_2, respectively. The extinction

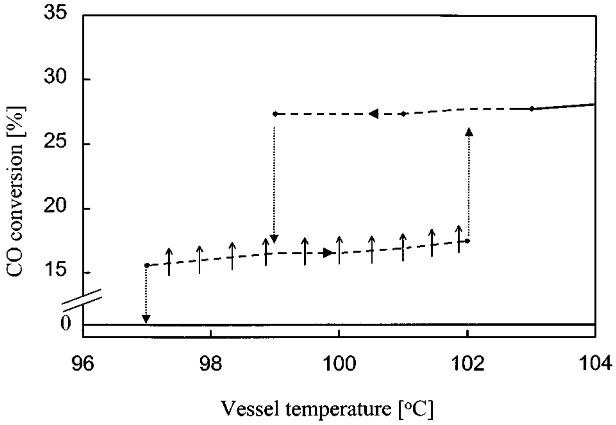

Figure 6. Dependence of CO conversion on vessel temperature for a feed consisting of 2.5, 28.8 and 68.7 vol. % of CO, O_2 and N_2, respectively.

Total feed rate of 3,880 cm^3/min. Single-headed arrows describe the oscillations amplitude of the effluent CO concentration of states with a hot zone and single-front motion.

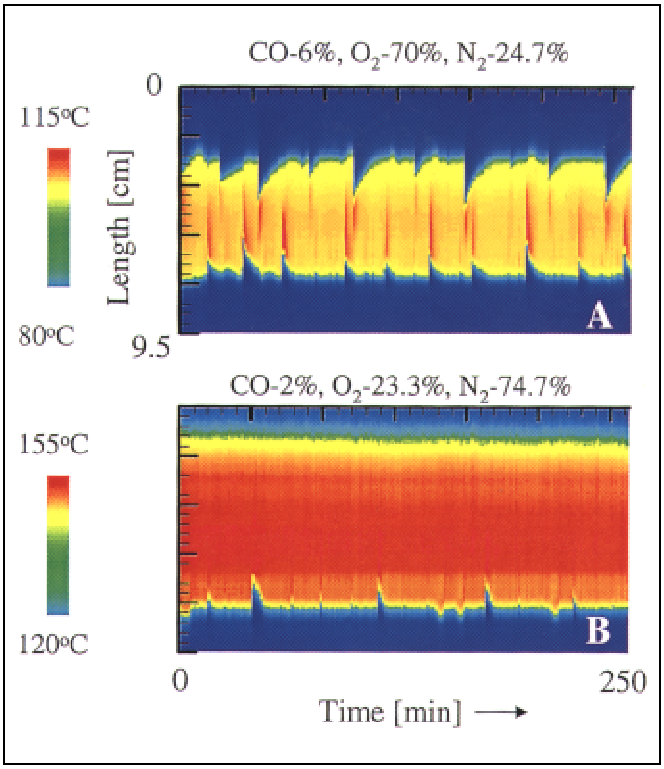

Figure 7. Two qualitatively different hot zones obtained for different feed compositions at a total flow rate of 1,200 cm^3/min.

(a) Hot zone with dual-front motion; (b) a hot zone with single-front motion.

temperature for the dilute feed (Figure 6) was higher than that for the richer feed mixture (Figure 3). Moreover, while the transition from the stationary to the oscillatory nonuniform states was smooth for the richer feed mixture (Figure 3), the transition from the stationary to oscillatory nonuniform states was abrupt for the diluted mixture (Figure 6). That transition between the branches of stationary nonuniform to oscillatory states generated a hysteresis loop. Thus, for some vessel temperatures, either a stable stationary nonuniform state or a stable oscillatory nonuniform state was obtained for the same feed and operating conditions. The initial conditions determined which of the two states was reached. The difference between the two nonuniform states is not just the motion or lack of motion of the temperature fronts, but a large difference in the size of the hot region and the corresponding time-average reaction rate.

While both temperature fronts surrounding the hot zone moved back-and-forth for the richer feed (Figure 7a), only one of the two fronts surrounding the hot zone moved back-and-forth for the more diluted feed (Figure 7b). It should be noted that while both hot zones were obtained for the same total flow rate of 1,200 cc/min and O_2/CO ratio of 11.66, the hot zones were attained at different vessel temperatures. The hot zone for the reaction mixture with 2 vol. % CO (Figure 7b) was attained at 105°C, close to the extinction temperature of 103°C. The hot zone for the feed with 6 vol. % CO was obtained at a vessel temperature of 62°C, close to the extinction temperature of 60°C. Thus, the difference in the dynamic features of the fronts bounding these hot zones may be due to the difference in the temperatures rather than that in the reactant concentrations. The qualitative features of the hot zone motions shown in Figure 7 were independent of the azimuthal position as long as the two cold regions next to the reactor ends did not coalesce and completely surround the hot zone.

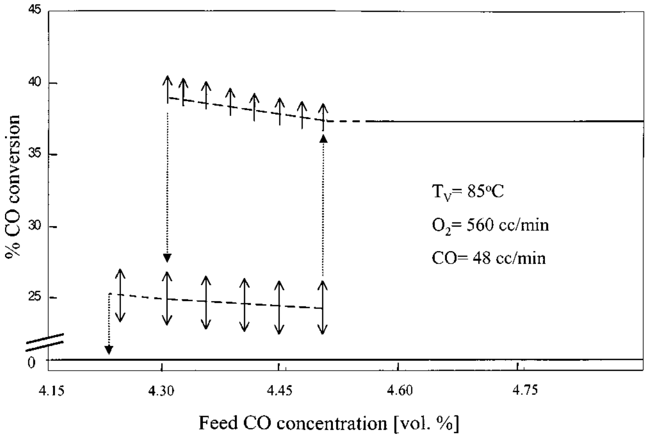

Figure 8. Dependence of CO conversion on CO feed concentration.

Total flow rate of O_2 and CO was 608 cm^3/min, and vessel temperature was 85°C.

In Figure 8, the bifurcation diagram of CO conversion vs. the limiting reactant concentration (conducted for a CO/O_2 ratio of 1/11 and a constant O_2 and CO flow rate of 608 cc/min and vessel temperature of 85°C) shows that two branches of stable states with qualitatively different hot zones existed over a bounded range of CO concentrations. One of the branches contains a relatively large hot zone, with only one of the two temperature fronts moving back-and-forth. The second branch consists of states with a smaller hot zone and both temperature fronts moving back-and-forth. A corresponding large difference exists in the conversion attained by the two different states. The initial conditions determine which of the two states is attained. The two branches generate a small hysteresis loop. At 4.50 vol. % CO, a slow *increase* of the CO concentration caused a transition (bifurcation) from the branch with the small hot zone to that with the larger hot zone and conversion. A slow *decrease* in the CO concentration at 4.33 vol. % CO caused a transition from the branch with the large hot zone to that with the smaller hot zone and lower conversion. Upon further decrease of the CO concentration, that hot zone extinguished at CO concentration of 4.23 vol. %.

Figure 9 illustrates the qualitative differences between the two states with a hot zone, existing for the same operating conditions and feed. For the sake of clarity, only a 4 cm section containing the hot zone in this length vs. time temperature map is shown. Note that in this case only one of the temperature fronts which bounds the wider hot zone (Figure 9a) is moving back-and-forth. In contrast, both temperature fronts bounding the smaller hot zone moved back-and-forth (Figure 9b).

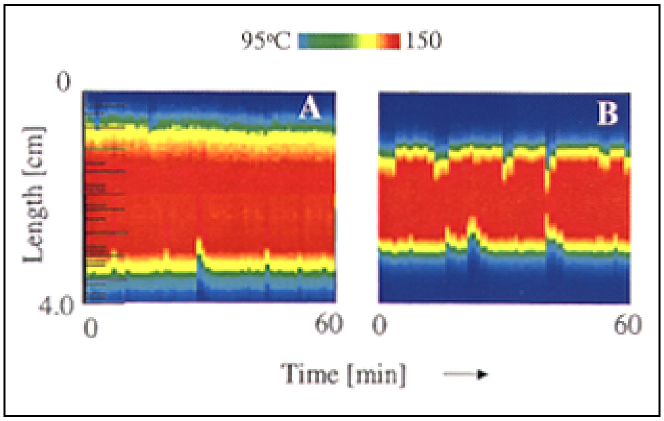

Figure 9. Two types of hot zones obtained for the same feed (CO-4.3 vol. %, O_2-50.5%, N_2-45.2%) and operating conditions.

Vessel temperature 85°C, Total feed rate of 1,100 cm^3/min.

Discussion and Conclusions

The existence of hot zones in a radial flow reactor can have a deleterious impact on the yield of the desired product. Moreover, it can initiate undesired reactions and lead to safety hazards. Boreskov et al. (1981) pointed out that transversal hot zones may form due to nonuniform packing of a packed-bed reactor. This effect was not important in the experiments in this article, as the reticulated support forming the radial flow reactor had a very uniform structure. Nguyen and Balakotaiah (1994) and Benneker et al. (1998) suggest that the interaction between free convection and reaction may generate hot and cold zones in a packed-bed reactor. The experiments did not reveal any difference in the hot region on the top and bottom of the horizontal held radial flow reactor. Thus, natural convection could not have been the cause of the hot region and its motion. Moreover, the dispersion of the gaseous reactants in the dense reticulated alumina structure is not likely to exceed the radial heat dispersion. Thus, the hot regions cannot be predicted by the psuedo-homogeneous model used by Balakotaiah et al. (1999). The reactor hot zone was generated by the interaction from the rate of the heat release by the chemical reaction, the removal of heat from the reactor, transversal heat conduction, and from the impact of the change in fluid properties (such as viscosity and density) on the flow field. In addition, the hot zones were affected to some extent by the inherent nonuniform catalytic activity of the impregnated reactor. The interaction among the various rate processes in this system is more intricate than in a diffusion-reaction system, where the impact of convection usually does not have to be taken into account.

The hot zones were much larger than the typical size of a catalytic pellet. This suggests that the evolution and dynamics of hot zones in a packed-bed reactor are governed by a different cause than those forming spatiotemporal patterns on individual catalyst pellets. In all the experiments in this work the velocity of the temperature fronts was very slow (order of mm/min), and the frequency of the back-and-forth motion was rather low. This is due to the high heat capacity of the bed relative to the rate of heat generation per unit volume of the reactor. A faster response is expected to exist for a reactor with a smaller volumetric heat capacity and a higher rate of heat generation.

The hot regions were always found for operating conditions close to the extinction of the high conversion, ignited branch. This is probably caused by high sensitivity of the reactor to the operating conditions near the extinction temperature. This sensitivity amplifies the impact of any nonuniformity. Our experiments suggest that hot zones are likely to form in a packed-bed reactor operating close to its extinction temperature. This conclusion is in agreement with the fact that Pt-Rh gauze converters, used in the synthesis of HCN and NO and which are run close to the extinction temperature to minimize the loss of the precious Pt-Rh, are known to have small, moving hot spots (flickering).

A novel and surprising finding of this study is that in some cases two disjoint, stable states exist with qualitatively different hot zones for the same feed and operating conditions. For example, Figure 6 illustrates the existence of two states with qualitatively different hot zones in a bounded range of vessel temperatures, while Figure 8 shows that two stable states with qualitatively different hot zones exist over a bounded range of CO concentration. The difference in the size and features of the temperature fronts bounding the hot zone is shown in Figure 9. The finding of two stable branches of states with qualitatively different hot zones raises the question as to whether an even more intricate hysteresis may exist, which is hidden and inaccessible by our experiments. There may exist other disjoint branches of nonuniform states which are totally nested within the overall hysteresis loop. These branches would not have been found by the experimental procedure used, and their experimental finding requires some−presently unavailable−theoretical guidance and ability to predict their existence. It is of practical importance and academic interest to determine if and under what conditions these branches may exist. A related important question concerns what is the number of different nonuniform states that may form in a multireaction system, which has more than two stable steady states under the same operating conditions (Harold and Luss, 1987).

Horvath et al. (1993) and Hagberg and Meron (1994) have shown that, in a two-variable diffusion-reaction system, an increase in the ratio of the time-constant of the inhibitor to that of autocatalytic variable can destabilize stationary fronts. In this system, the limiting reactant concentration is the inhibitor and its time constant is (1/(the pseudo first-order reaction rate constant [k]). The time constant of the temperature, which is the autocatalytic variable, depends on the transport coefficients and physical properties and is rather insensitive to changes in the temperature. We noted in all our experiments that a decrease in the vessel temperature, which implies an increase in the time constant of the limiting reactant, tended to destabilize the temperature front and increase the frequency and amplitude of its motion. This trend is in agreement with the predictions of the Hagberg and Meron (1994) analysis.

A dilution of the limiting reactant in the feed shifted the extinction temperature to higher vessel temperatures. This explains the transition from uniform states to those with a hot zone upon dilution of the feed at a constant vessel temperature, as shown in Figure 8. The dilution of the limiting reactant shifted the reactor closer to its extinction temperature, next to which the hot zones tend to appear. For the higher feed concentration, no hot zones formed as their temperature was much higher than the extinction temperature.

The two fronts bounding the hot zone did not move in tandem, and their back-and-forth motion had different amplitudes. This is probably due to the nonuniformity of the catalytic activity of the reactor, and the inefficient lateral dispersion of heat between the two fronts. For the high reactant concentrations, both temperature fronts bounding the hot zone moved back-and-forth. In contrast, in some of the experiments with the more dilute feeds, only one of the two fronts moved back-and-forth. This is most likely due to the lower temperatures at which the hot zone formed for the more concentrated feed. As mentioned, the time constant of the inhibitor tends to increase with decreasing temperature and to destabilize stationary fronts.

The stationary temperature front was always generated at the same side of the reactor. Moreover, when the reactor was turned around between the two supports, the stationary temperature front moved to the other side of the vessel. This supports the claim that some nonuniformity in the catalytic activity of the reticulated structure was responsible for this asymmetric behavior. Any nonuniformity in the flow field caused by the flow distributor or quartz sleeve would not have shifted the front location upon turning the reactor around between the two supports. Inhomogeneities in the catalytic activity are inherent features of heterogeneous catalytic systems (Philippou et al., 1991; Pawlicki and Schmitz, 1987; Kellow and Wolf, 1991; Annamalai et al., 1997, 1999; Liauw et al., 1996; Somani et al., 1997). Moreover, surfaces that initially have a uniform activity will change with time and become nonuniform (induced nonuniformity) under atmospheric or higher pressures. Experiments with single catalytic pellets indicate that this nonuniformity affects the shape and location of either the hot or cold regions on the surface of a single pellet. Clearly, these nonuniformities are likely to affect the evolution and dynamic features of hot zones in packed-bed reactors.

Several previous studies reported temperature (Puszynnski and Hlavacek, 1984) or concentration (Rovinsky and Menzinger, 1993) pulses moving in the flow direction in packed-bed reactors. Similarly, there have been simulations of complex pulse motion in the flow direction (Sheintuch and Nekhamkina, 1999). The experiments in this work are of transversal hot zones forming on a surface normal to that of the flow. The only other experimental report of such transversal hot zones in a packed-bed reactor is that by Boreskov et al. (1981). It was generated by nonuniform packing of the bed. A theoretical analysis of transversal temperature nonuniformities was recently presented by Balakotaiah et al. (1999). However, their model cannot predict the hot zone formation when the radial dispersion of heat exceeds that of the species dispersion (Yakhnin and Menzinger, 2001). A remaining important concern is the impact of the reactor cross-section size on the type and number of transversal hot zones which may form. This requires discovering if the hot zones have a characteristic size which is independent of the size of the system, as is the case in many diffusion–reaction systems, or if their size is related to that of the reactor and pellets. Previous studies of different states on the two faces of a slab catalyst (Pismen et al., 1968; Luss et. al., 1972) suggest that different transversal hot zones can co-exist when the characteristic time of heat conduction between the two exceeds by some critical value that of the local heat generation. For a pseudo first-order reaction, this implies that $L^2k(T)/\alpha$ has to exceed some critical value. Here, α is the transverse effective heat conductivity, and L is the distance between the two hot zones, and $k(T)$ is the pseudo-first-order rate constant.

Acknowledgments

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Literature Cited

Annamalai, J., C. Ballandis, M. Somani, M. A. Liauw, and D. Luss, “Effects of Reactant Composition and Nonuniformities on Temperature Fronts,” *J. Chem. Phys.*, **107**, 1896 (1997).

Annamalai, J., M. A. Liauw, and D. Luss, “Temperature Patterns on a Hollow Cylindrical Catalyst Pellet,” *Chaos*, **9**, 36 (1999).

Balakotaiah, V., E. L. Christoforatou, and D. H. West, “Transverse Concentration and Temperature Nonuniformities in Adiabatic Packed-Bed Catalytic Reactors,” *Chem. Eng. Sci.*, **54**, 1725 (1999).

Barkelew, C. H., and B. S. Gambhir, “Stability of Trickle-Bed Reactors,” *ACS Symp. Ser.*, **237**, 61 (1984).

Benneker, A. H., A. E. Kronberg, and K. R. Westerterp, “Influence of Buoyancy Forces on the Flow of Gases through Packed Beds at Elevated Pressures,” *AIChE J.*, **44**, 263 (1998).

Boreskov, G. K, Y. S. Matros, O. P. Klenov, V. I. Lugovskoi, and V. S. Lakhmostov, “Local Nonuniformities in a Catalyst Bed,” *Dokl. Akad. Nauk SSSR*, **258**, 1418 (1981).

Colin, P., and V. Balakotaiah, “Analysis of Concentration and Temperature Patterns on Catalytic Surfaces,” *J. Chem. Phys.*, **100**, 5338 (1994).

Field, R. J. and M. Burger, eds., *Oscillations and Travelling Waves in Chemical Systems*, Wiley, New York (1985).

Hagberg, A., and E. Meron, “Pattern Formation in Non-Gradient Reaction-Diffusion Systems: The Effects of Front Bifurcations,” *Nonlinearity*, **7**, 805 (1994).

Harold, M. P., and D. Luss, “Use of Multiplicity Features for Kinetic Modeling: Carbon Monoxide Oxidation on Platinum-Alumina,” *Ind. Eng. Chem. Res.*, **26**, 2099 (1987).

Horvath, D., Petrov, V., Scott, S. K., and K. Showlater, “Instabilities in Propagating Reaction–Diffusion Fronts,” *J. Chem. Phys.*, **98**, 6332 (1993).

Jaffe, S. B., “Hot Spot Simulation in Commercial Hydrogenation Processes,” *Ind. Eng. Chem. Proc. Des. Dev.*, **15**, 410 (1976).

Kellow, J. C., and E. E. Wolf, “Propagation of Oscillations During Ethylene Oxidation on a Rh/SiO_2 Catalyst,” *AIChE J.*, **37**, 1844 (1991).

Lee, J. P., V. Balakotaiah, and D. Luss, “Steady-State Multiplicity of a Packed-Bed Reactor Operating Under a Fixed Pressure Drop,” *Chem. Eng. Sci.*, **41**, 749 (1986).

Liauw, M. A., J. Ning, and D. Luss, “Pattern Formation on a Nonuniformly Active Ring,” *J. Chem. Phys.*, **104**, 5657 (1996).

Luss, D., J. E. Bailey, and S. Sharma, “Asymmetric Steady States in Catalytic Slabs with Uniform and Nonuniform Environments,” *Chem. Eng. Sci.*, **27**, 1555 (1972).

Marwaha, B., J. Annamalai, and D. Luss, “Hot Zone Formation during Carbon Monoxide Oxidation in a Radial Flow Reactor,” *Chem. Eng. Sci.*, **56**, 89 (2001).

Matros, Yu. Sh., and N. A. Chumakova, “Multiplicity of Stationary Regimes in an Adiabatic Catalyst Layer,” *Dokl. Akad. Nauk USSR.*, **250**, 1421 (1980).

Nguyen, D., and V. Balakotaiah, “Flow Mal-Distributions and Hot Spots in Down-Flow Packed-Bed Reactors,” *Chem. Eng. Sci.*, **49**, 5489 (1994).

Pawlicki, P. C., and R. A. Schmitz, “Spatial Effects on Supported Catalysts,” *Chem. Eng. Prog.*, **83**, 40 (1987).

Philippou, G., F. Schultz, and D. Luss, “Spatiotemporal Temperature Patterns on a Electrically Heated Catalytic Ribbon,” *J. Phys. Chem.*, **95**, 3224 (1991).

Pismen, L. M., and Y. L. Kharakats, “Asymmetric State of a Heterogeneous Exothermic Reaction,” *Dokl. Akad. Nauk SSSR*, **178**, 901 (1968).

Puszynski, J., and V. Hlavacek, “Experimental Study of Ignition and Extinction Waves and Oscillatory Behavior of a Tubular Nonadiabatic Packed-Bed Reactor for the Oxidation of Carbon Monoxide,” *Chem. Eng. Sci.*, **39**, 681 (1984).

Rovinsky, A. B., and M. Menzinger, “Self-Organization Induced by the Differential Flow of Activator and Inhibitor,” *Phys. Rev. Lett.*, **70**, 778 (1993).

Sheintuch, M., and O. Nekhamkina, “Pattern Formation in Homogeneous and Heterogeneous Reactor Models,” *Chem. Eng. Sci.*, **54**, 4535 (1999).

Somani, M., M. A. Liauw, and D. Luss, “Evolution and Impact of Temperature Patterns during Hydrogen Oxidation on a Ni Ring,” *Chem. Eng. Sci.*, **52**, 2331 (1997).

Yakhnin, V., and M. Menzinger, “On Transverse Patterns in Packed-Bed Catalytic Reactors,” *Chem. Eng. Sci.*, **56**, 2233 (2001).

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