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Experimental Study of Temperature Profiles in a Rapidly Deactivating Catalyst Bed

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Temperature profiles and exit conversion have been studied for different inlet poison concentrations, overall amount of poison fed to the reactor, and inlet concentration of the key component. The catalyst used in this study was industrial-type Pt–Al_2O_3 (approx 0.1 wt% Pt), and the reaction conducted was the CO oxidation. CS_2 was used as catalyst poison, and the deactivation process was reversible. It was observed experimentally that, during the deactivation process, excessive hot spots occurred which moved downstream. Qualitative agreement between the observed results and those predicted from theory was obtained.

INTRODUCTION

Catalyst deactivation is of major concern in catalyst development and design of packed-bed reactors. The interaction of the chemical and physical processes taking place in a porous catalyst may give rise to unexpected trends; for instance, recently Ervin and Luss (*1*) and Blaum (*2*) have theoretically predicted that, for an exothermic reaction, the temperature in the bed in the course of the deactivation process may greatly exceed the adiabatic temperature.

So far there are only a few papers that have been published that give experimental measurements of temperature and concentration profiles in a deactivating packed bed where an exothermic reaction occurs (*2–8*). In all these measurements, with the exception of the work of Blaum (*2*), the value of the hot spot temperature decreases as a result of a particular deactivation process.

The main objective of this paper is to obtain experimental information on the hot spot temperature in a poisoned fixed bed and to support the theoretical conclusions of Luss (*1*) and Blaum (*2*) with experimental observations.

EXPERIMENTAL

Oxidation of CO in a deactivating bed was studied in an adiabatic tubular reactor provided with an evacuated jacket covered by a silver coating (cf. Fig. 1). The characteristics of the reactor and catalyst used are presented in Table 1 and Table 2.

The reactor temperature was measured by an iron–constantan thermocouple placed in an axial thermowell. Since the value of the overall heat transfer coefficient was lower than 10^{-3} kcal/m^2 sec °K and the reactor diameter was small, the radial temperature gradients have not been considered. The value of the overall heat transfer coefficient was determined experimentally from measurements performed in the absence of a chemical reaction; this value was confirmed also by calculations considering all radial thermal resistances.

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TABLE 1

Characteristics of Reactor and Catalyst

Reactor length	0.56 m
Reactor internal diameter	0.026 m
Reactor external diameter	0.060 m
Catalyst bed length	0.16 m
Catalyst	$Pt-Al_2O_3$, spherical particles
Catalyst diameter	0.0034 m

The poison, CS_2, was injected as a liquid in a preheated gas stream (CO, O_2), and immediate evaporation resulted. The rate

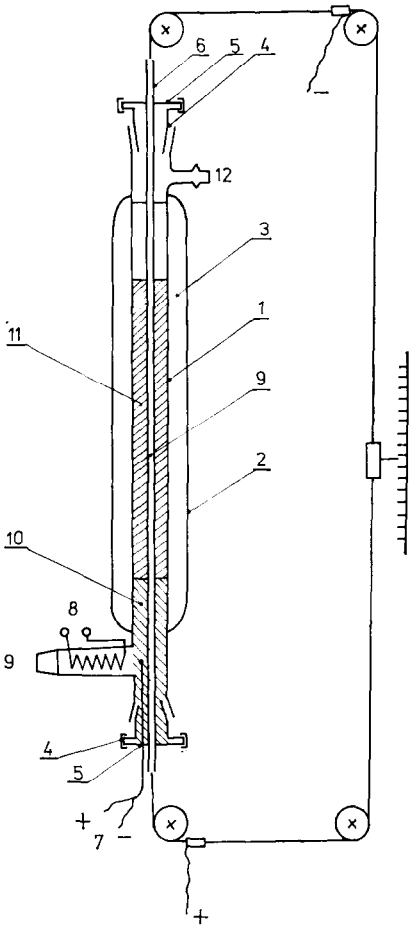

FIG. 1. Laboratory tubular adiabatic reactor: (1) internal tube, (2) external tube, (3) evacuated jacket with silver coating, (4) ground joints, (5) silicon rubber packing gland, (6) thermowell, (7) scale, (8) auxiliary preheater of inlet gas, (9) inlet of reacting gas, (10) glass beads, (11) catalyst, (12) gas exit.

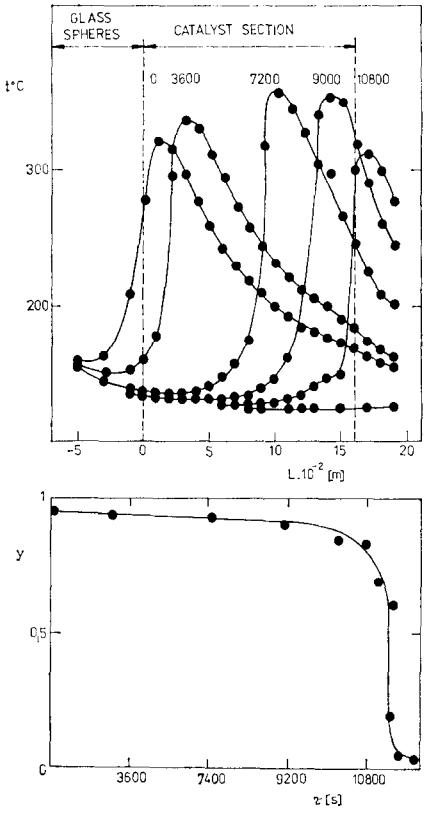

FIG. 2. Transient temperature and conversion profiles in the bed. Continuous deactivation. Inlet CO concentration: 3% CO; inlet poison concentration: 0.0063% CS_2; $G = 3.95 \cdot 10^{-5}$ kg/sec.

of deactivation, i.e., the poison concentration, was selected in such a way that the temperature profile did not change during measurements (approx 60 sec). The composition of the exit gas was analyzed by a thermal conductivity cell.

TABLE 2

Characteristic Operating Conditions and Ranges for the Variables

Volumetric flow rate (standard conditions)	1.78–1.9 liter/min
Linear flow rate (interstitial, standard conditions)	13.9 –14.9 cm/sec
Inlet CO concentration	1 or 3%
Inlet temperature	150°C
N_{Re}	9.6

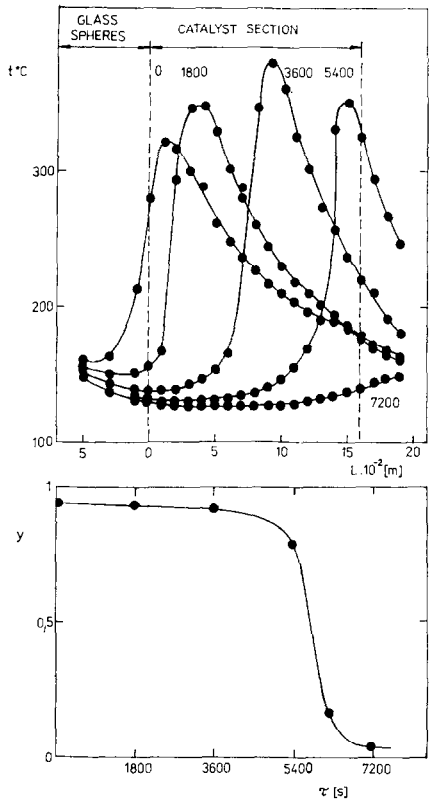

FIG. 3. Transient temperature and conversion profiles in the bed. Continuous deactivation. Inlet CO concentration: 3%; inlet poison concentration: 0.023% CS_2; $G = 3.95 \cdot 10^{-5}$ kg/sec.

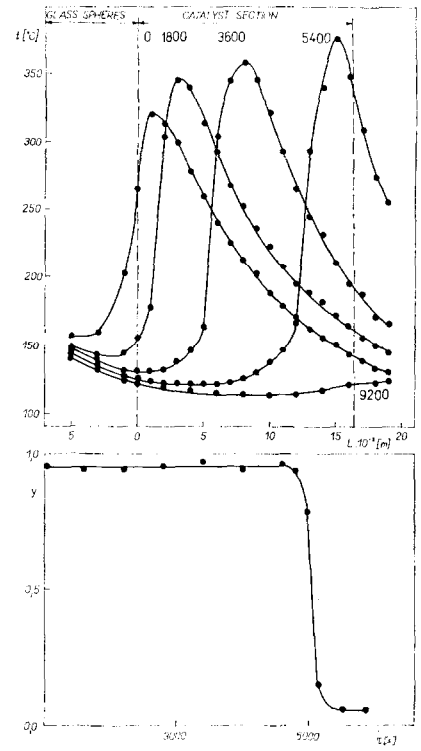

FIG. 4. Transient temperature and conversion profiles in the bed. Continuous deactivation. Inlet CO concentration: 3%; inlet poison concentration: 0.0339% CS_2; $G = 3.95 \cdot 10^{-5}$ kg/sec.

RESULTS AND DISCUSSION

(a) *Reactor Behavior for a Continuous Deactivation*

The effect of the inlet poison concentration on reactor dynamics may be inferred from Figs. 2–4. It is obvious that the progress of the hot spot depends on the inlet poison concentration; the higher the feed poison concentration the higher the speed of the moving zone. Similar behavior also may be observed in Figs. 5 and 6. Notice that the rate of catalyst deactivation is higher at low temperatures; this observation is in accordance with the deactivation mechanism proposed since, at higher temperatures, oxidation of CS_2 occurs. The speed of the hot spot zone is

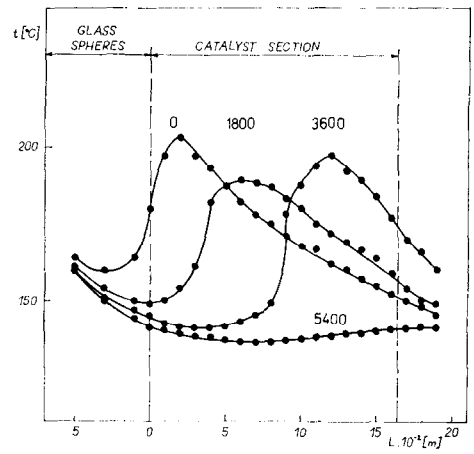

FIG. 5. Transient temperature profiles in the bed. Continuous deactivation. Inlet CO concentration: 1%; inlet poison concentration: 0.0063% CS_2; $G = 4.19 \cdot 10^{-5}$ kg/sec.

time-dependent. At first, the deactivation process occurs slowly because of the high inlet temperature which results in the combustion of CS_2. In the course of the process, the preheating effect of the dead inlet zone gives rise to a higher speed for the hot spot region. For lower values of inlet poison concentration, the hot spot temperature reaches a maximum value and then slowly decreases (see Figs. 2 and 3), while for higher poison concentrations, the hot spot temperature increase continuously (cf. Fig. 4).

(b) *Reactor Behavior for a Discontinuous Deactivation*

The effect of the integral amount of poison fed is shown in Figs. 7, 8 and 9. For a higher inlet poison concentration, the poison was fed for a shorter period. Evidently, in the first period, oxidation of the poison occurs, i.e., the amount of the poison adsorbed is low, and, if the poison inlet concentration decreases to zero, only a small amount of the poison can migrate downstream. As a result, the deactivation process is slower. This conclusion has been verified

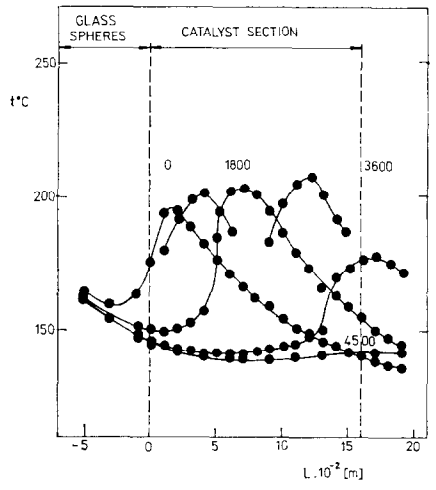

FIG. 6. Transient temperature profiles in the bed. Continuous deactivation. Inlet CO concentration: 1%; inlet poison concentration: 0.023% CS_2; $G = 4.19 \cdot 10^{-5}$ kg/sec.

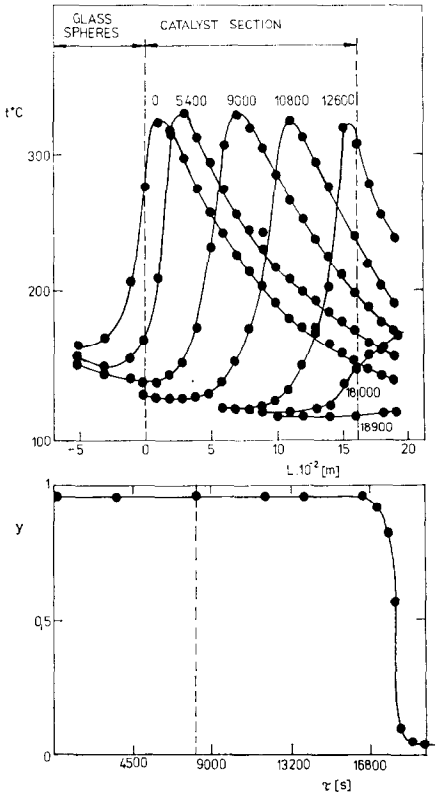

FIG. 7. Transient temperature and conversion profiles in the bed. Discontinuous deactivation. Deactivation process stopped after 8100 sec. Inlet CO concentration: 3%; inlet poison concentration: 0.0063% CS_2; $G = 3.95 \cdot 10^{-5}$ kg/sec

experimentally; when the total amount of poison was fed as a Dirac function, catalyst deactivation was not observed. The speed of the hot spot zone is slower in comparison to the case shown in Figs. 2–4 since the deactivation process is caused only by poison migration adsorbed in the front part of the reactor.

The course of the reactor extinction and ignition can be observed in Fig. 8. If the poison feed concentration decreases to zero, then the hot spot temperature also decreases, and, finally, an extinction process occurs. This process is then followed by a slow increase in temperature in the whole reactor, since the majority of poison is desorbed. Finally, this temperature increase is associated with the reactor ignition

which occurs in the inlet part of the reactor.

To illustrate the effect of the poison adsorbed in the inlet section, the feed of poison was stopped if the hot spot reached a certain position in the reactor. For different feed poison concentrations, different amounts adsorbed in the same inlet section. If the given state has been attained by a reacting gas with higher inlet poison concentration, then the time necessary for reactor extinction is less (cf. Figs. 7 and 8).

CONCLUSIONS

Based on the experimental observations, the following conclusions can be drawn. (i) During the deactivation process the hot spot zone moves downstream. (ii) The speed of the hot spot is not constant. The slowest progress occurs at the beginning of the deactivation process, and progress accelerates toward the reactor outlet. (iii) The temperature in the hot spot increases in accordance with the rate of deactivation. (iv) Deactivation by CS_2 is reversible if an oxidation reaction occurs. After decreasing the inlet poison concentration to zero, poison desorption occurs, and the activity of the catalyst bed is regenerated. (v) For a lower inlet poison concentration, deactivation of the bed requires a smaller integral amount of poison.

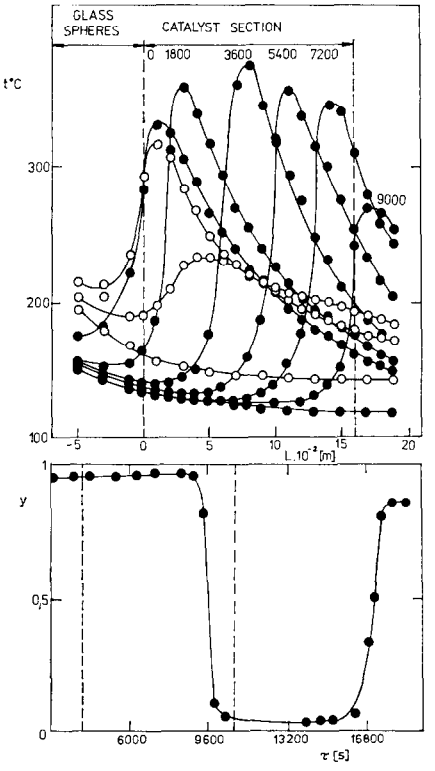

FIG. 8. Transient temperature and conversion profiles in the bed. Discontinuous deactivation. Deactivation process stopped after 3060 sec. Inlet CO concentration: 3%, inlet poison concentration: 0.023% CS_2; $G = 3.95 \cdot 10^{-5}$ kg/sec.

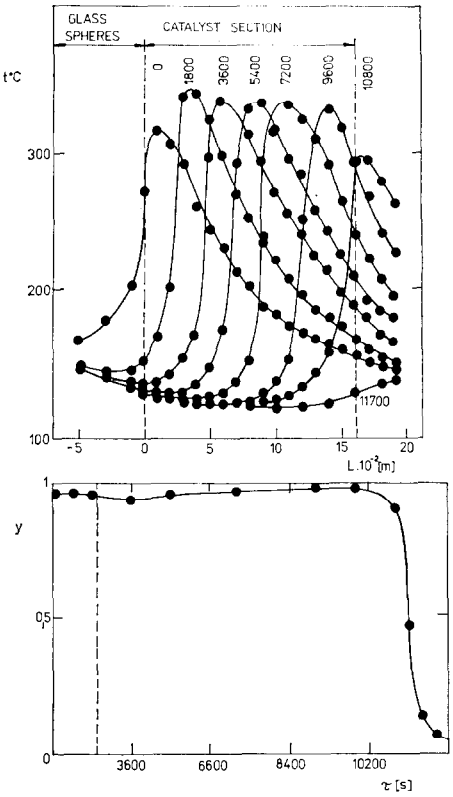

FIG. 9. Transient temperature and conversion profiles in the bed. Discontinuous deactivation. Deactivation process stopped after 1980 sec. Inlet CO concentration: 3%; inlet poison concentration: 0.0339% CS_2; $G = 3.95 \cdot 10^{-5}$ kg/sec.

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