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NOTES

Experimental and Theoretical Testing of a Perfectly Mixed Continuous Microreactor

A very attractive catalytic continuously stirred tank microreactor operating at atmospheric pressure was developed a few years ago by Mencier *et al.* (*1*). The essential feature of this device is that mixing is realized by superimposing a reciprocating flow to the main flow. The backward and forward motion of the fluid is due to two membrane compressors set up in opposition and located on both sides of the catalytic bed (see Fig. 1). The main component of the apparatus is a synchronous buzzer, i.e., one for which frequency is fixed by the electric current frequency. This buzzer puts in motion 100 times per second the Teflon membrane of each compressor by successive attraction and repulsion (50-Hz-frequency alternating current). The amplitude of the membrane motion is controlled by the voltage of the current. High reciprocating to main flow ratios are easily obtained.

Considering the working principle of this type of reactor, Mencier *et al.* (*1*) demonstrated perfect mixing with regard to composition by verifying equality of compositions above and below the bed, in the case of the catalytic dehydrogenation of cyclohexane. We have also used with success such a chemical test with the catalytic hydrogenation of ethylene (*2*).

The objective of the present study is to demonstrate the perfect mixing of this reactor by a general physical test based on the stimulus–response technique (*2*).

The experimental system is represented in Fig. 2. The reactor is supplied with hydrogen at a perfectly constant flow rate. The tracer input circuit (argon in our case) comprises two electromagnetic valves allowing the reproducible injection of very small gaseous volumes. Two three-way stopcocks V_1 and V_2 allow one to bypass the microreactor (its volume is equal to 1 cm^3) and the two membrane compressors set on both sides. From the injection point, capillary pipes are used (inside diameter equal to 2 mm) in order to approach plug flow. Thermal conductivity fluctuations of gas-

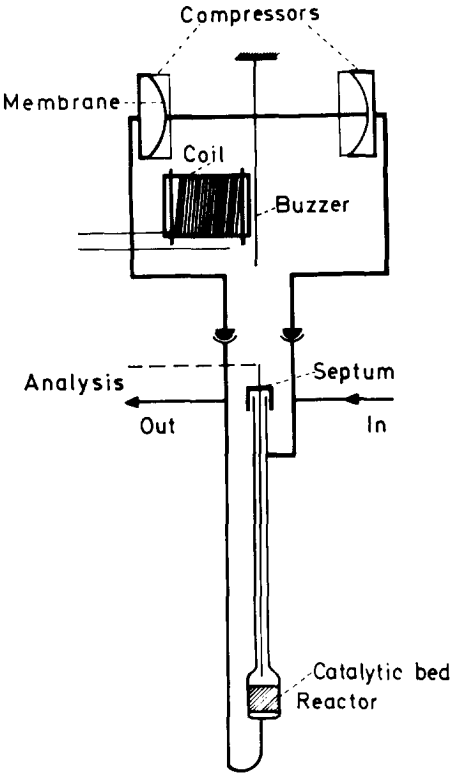

FIG. 1. Scheme of the reactor.

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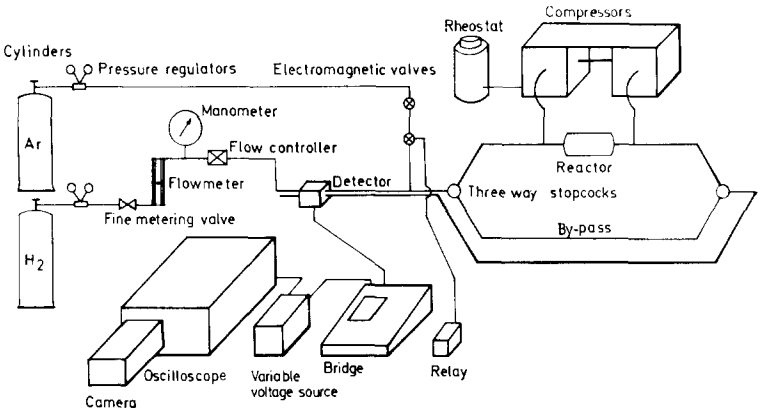

FIG. 2. Apparatus for the stimulus–response study.

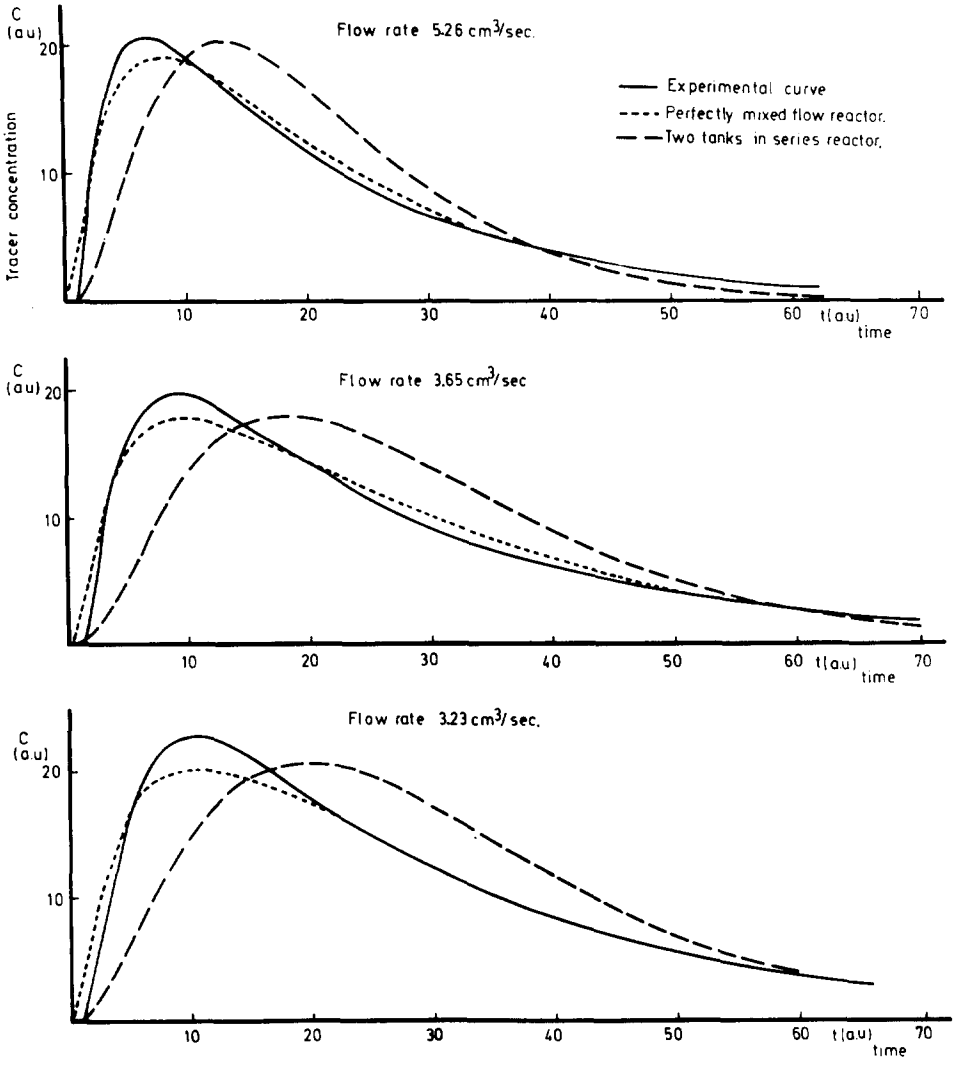

FIG. 3. Experimental and theoretical responses for three different flow rates.

TABLE 1

Transfer Functions in Laplace Space: Results of Adjustments[a]

Flow rate (cm^3/sec)	n Tanks in series model (n)	Axial dispersion model (Pe)
3.23	1.02	0.25
3.65	0.90	0.08
5.26	0.85	0.00

[a] (i) n Tanks in series model:

$$H(p) = \left(1 + \frac{p}{n}\right)^{-n},$$

where p = complex variable and n = number of tanks. (ii) Axial dispersion model with Danckwerts boundary conditions:

$$H(p) = \frac{4[1 + (4p/\mathrm{Pe})]^{\frac{1}{2}} e^{\mathrm{Pe}/2}}{\{1 + [1 + (4p/\mathrm{Pe})]^{\frac{1}{2}}\}^2 e^{(\mathrm{Pe}/2)[1+(4p/\mathrm{Pe})]^{\frac{1}{2}}} - \{1 - [1 + (4p/\mathrm{Pe})]^{\frac{1}{2}}\}^2 e^{-(\mathrm{Pe}/2)[1+(4p/\mathrm{Pe})]^{\frac{1}{2}}}},$$

where $\mathrm{Pe} = V \cdot L/D_L$ = Peclet number, V = fluid velocity through the vessel (m/sec), L = reactor length (m), D_L = effective axial dispersion coefficient (m^2/sec).

eous flow are detected by a catharometer with two filaments for microanalysis. Signals are recorded on an oscilloscope screen and photographed. This experimental system allows us to measure residence times of less than 1 sec.

The study of flow through a vessel by the stimulus–response technique consists of disturbing the system and then seeing how it responds to the stimulus. In our problem, the stimulus is a tracer input into the fluid entering the vessel, whereas the response is a time record of the tracer leaving the vessel. Comparison between input and output allows one to determine the macromixing of the system.

The reactor transfer function in Laplace space can be written as

$$E(p) = \frac{M_1(p)}{M_2(p)} \cdot \frac{I_2(p)}{I_1(p)}, \qquad (1)$$

where M_1 = measurement of the reactor response to a tracer input I_1 (=the response), and M_2 = measurement of the bypass response to a tracer input I_2 (=the signal).

If the shapes of inputs I_1 and I_2 are identical, the ratio between the two normalized responses allows one to obtain the Laplace transform of the reactor function. Equation (1) is valid if the flows through the capillary pipes at the reactor entrance and exit and in the bypass are near plug flow. In this case, tracer inputs are not distorted, but only delayed.

The experimental transfer function is obtained by calculating the ratio between the Laplace transforms of the two previously normalized responses in p points of the Laplace plan.

The theoretical transfer functions $H(p)$ have been fitted on the experimental transfer functions $E(p)$ by simultaneous adjustment of the real and imaginary parts. The adjustment has been made on the positive part of the imaginary axis of the Laplace complex plan (*3*).

The responses to the experimental inputs of simple theoretical models such as the perfectly mixed flow model and the two tanks in series model have been calculated by decomposition of the input in rectangles and summation of the individual responses.

The reactor has been studied in the flow rate range between 0 and 19 liters/hr.

The experimental response and the theoretical responses of the perfectly mixed flow

reactor and of the two tanks in series reactor at three different flow rates are shown in Fig. 3. It can be seen that the perfectly mixed flow reactor model satisfyingly represents the experimental reactor behavior.

To conclude, transfer functions of simple parametric models, namely, the n tanks in series model and the axial dispersion model with Danckwerts boundary conditions (*4*), have been adjusted on the experimental transfer functions. These models approach the perfectly mixed flow reactor when the number of tanks goes to unity and the Peclet number to zero.

Results of adjustments given in Table 1 also allow us to conclude that the experimental reactor behaves like a continuously stirred tank reactor. Thus the chemical test proposed by Mencier *et al.* (*1*) and the physical test by the stimulus–response technique have shown that the experimental reactor presented can be simulated satisfyingly by the perfectly mixed flow model.

This reactor can, in some cases, be advantageously used instead of the differential reactor with external recycle, which has a similar mass balance equation. Its principal advantages are cheapness (~$100), strength, easily stabilized flow rate, and small dead zones.

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