

A Chemical Reaction Engineering Laboratory Experiment: Isothermal Laminar-Flow Reactor

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Abstract

We report here an experiment for the chemical reaction engineering laboratory. The reaction between sodium hydroxide and ethyl acetate is conducted isothermally in a tubular reactor under isothermal, laminar flow, conditions. Steady-state reactor performance is followed at different space times and Reynolds numbers. Analysis of reactor performance is easily followed by direct sampling and titration. Comparisons between theory and experiment are in reasonably good agreement. The experiment demonstrates how concepts in transport phenomena, introduced early in the chemical engineering curriculum, are related to reactor engineering.

Key words: reaction engineering laboratory; laminar flow reactor; kinetics.

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Introduction

The traditional approach in the undergraduate chemical reaction engineering (CRE) laboratory, as practiced in our university (and perhaps other universities) typically consists of experiments involving homogenous reactions in *closed* (batch) and *open* (continuous) tank and tubular reactors. In our CRE laboratory, batch-reactor experiments are used to collect concentration-time data using ex-situ and in-situ techniques, with the objective of determining reaction kinetics. Continuous tank- and tubular-reactor experiments are used to study transient and steady-state reactor performance.

Experiments on the tubular reactors, in particular, are normally operated under turbulent-flow conditions such that *plug flow* is a reasonable approximation of reactor flow conditions. This approach allows students to compare experimentally collected data with what is typically taught in the undergraduate CRE course. A typical CRE course mainly involves analysis of homogenous, single and multiple, reactions in *isothermal* and *non-isothermal*, “ideal” reactors. In our course, time limitations permit an elementary coverage of heterogeneous, solid-catalyzed, reactions and non-ideal homogeneous reactors. Attempts to include experiments on non-ideal reactors or solid-catalytic reactions in the CRE laboratory have been limited. Therefore we have been interested over recent years in introducing one or two new experiments that can widen the scope of the laboratory and offer instructors and different groups of students some practical options.

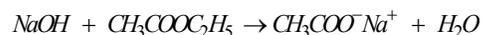
The objective of this communication is to summarize our experience with a variation of a classic experiment for the CRE laboratory. The experiment is intended to

introduce the student to the isothermal *laminar* flow reactor (LFR). While plug flow is a good approximation for most cases involving highly turbulent flows in long tubes, it is not for cases involving laminar flow. In the LFR, radial gradients in velocity, composition and temperature are significant, whereas in the *plug* flow reactor (PFR) the velocity profile is assumed to be flat. Thus in the LFR fluid properties vary in both axial and radial directions.

In doing this experiment, it is also hoped to counter-balance the common impression, often perceived by chemical engineering graduates, that industrial tubular reactors are only operated as PFRs. This approach, while still based on homogenous reactions, is also pedagogically advantageous, as it relates CRE to the first transport phenomena course that undergraduate students are introduced to early in the chemical engineering curriculum. Experiments on tubular reactor operation, in the laminar flow regime, were reported by Hovorka and Kendall (1960), and Kendall (1967). However, the emphasis in those studies was on effect of reactor geometry, and comparisons with theoretical results were limited.

Experimental

The system of interest in this experiment is the reaction between aqueous solutions of sodium hydroxide and ethyl acetate:



Kinetics of this well-known reaction are studied in the first experiment in the CRE laboratory using a batch reactor. Typical results of a kinetics experiment are given in the next section. The experimental set-up, used in the LFR experiment is manufactured by Armfield [www.armfield.co.uk/cex

_datasheet.html]. The set-up is shown in Figure 1.

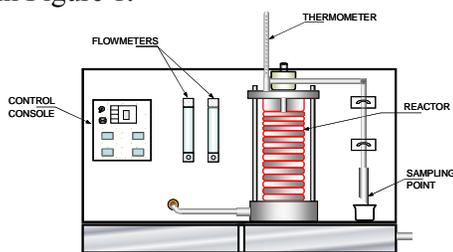


Fig. 1 Schematic of tubular reactor experimental set-up.

Solutions of the 0.1 M of caustic soda and 0.1 M of ethyl acetate are placed upstream in two storage tanks containing at least 20 lit of each solution. The tubular reactor, whose volume is 0.4 dm³ and inside diameter is 4 mm, is immersed in a thermostatted water bath. To ensure a homogeneous thermal field a continuous stirrer is also mounted within the water bath. In the experiments reported below, the temperature was maintained constant in the range 25-26 °C. Two flow meters were used to set the inflow of the two reagents. The reaction was carried out at different volumetric flow rates. The inlet flow rates of the two reactants were set and the total outlet flow rate was measured using a graduated cylinder and a stop watch. The sampling point is located at the outlet of the reactor. Samples of 2.5 dm³ from the reactor outlet were taken at different flow rates; quenched with an excess 0.05 M HCl solution; and back-titrated using 0.05 M sodium hydroxide solution after adding two to three drops of phenolphthalein. The volume of the sodium hydroxide solution needed to reach the end point (i.e. turn the color of the solution into stable pink) was recorded. Sampling at short time intervals was required to ensure that steady state was reached (usually in 4-5 minutes). Thus the

steady-state outlet concentration of sodium hydroxide was determined.

Results and Discussion

LFRs are open systems characterized by the continuous exchange of mass between the reactor boundaries and the environment. The flow in these reactors is characterized as laminar when the Reynolds number, $Re < 2100$. Laminar flow therefore occurs in situations characterized by low velocity, high viscosity, or small tube size.

In cylindrical tubes laminar flow is further characterized by a parabolic velocity profile (Wilkes, 1999); i.e. $u=u(r)$. Due to the absence of mixing in the axial (z) and the radial directions (r) in this reactor, concentration (and temperature) will vary in both directions, unlike the PFR where properties are presumed to vary only in the axial direction.

In the case of an isothermal, constant-density, irreversible reaction, whose rate is $(-R_A)$, the performance equation of the LFR is given by the following mole balance equation:

$$u(r) \frac{\partial C_A}{\partial z} + (-R_A) = 0 \quad (1)$$

In term of the mean residence time τ , this equation becomes:

$$\frac{\partial C_A}{\partial \tau} + (-R_A) = 0 \quad (2)$$

These equations apply at a particular radial position r ; therefore one must integrate radially to obtain an *average* concentration $\langle C_A \rangle$ at any axial position $z \in (0, L]$, where L is reactor outlet. Note that $C_A(L)$ is of interest here as concentration is measured at the outlet in this experiment. Solutions of equation (2) in the case of Newtonian fluids were given by Bosworth (1948), Cleland and

Wilhelm (1956), and Denbigh (1951), for zero-, first- and second-order reactions, respectively. These solutions are also reported in most standard CRE textbooks.

For a LFR of inside diameter D and length L , the average outlet concentration, in the case of second-order kinetics, where *equimolar* amounts of the reactants are used as in the present experiment, is given by:

$$\langle C_A \rangle = C_{A0} \left[1 - kC_{A0}\tau \left[1 - \frac{kC_{A0}\tau}{2} \ln \left(1 + \frac{2}{kC_{A0}\tau} \right) \right] \right] \quad (3)$$

Therefore conversion of reactant A (sodium hydroxide in our case) is given by (Missen et al., 1999):

$$\langle x_A \rangle = kC_{A0}\tau \left[1 - \frac{kC_{A0}\tau}{2} \ln \left(1 + \frac{2}{kC_{A0}\tau} \right) \right] \quad (4)$$

It is clear that use of Eq. (3) requires kinetics data for the reaction. As alluded to earlier, such data were collected in a batch reactor. Concentrations of sodium hydroxide versus time were utilized to determine the kinetics of the reaction under study. By using the integral method, in which the reaction rate expression is assumed and tested using experimental data, it was confirmed that the reaction is second-order and the reaction rate constant was calculated to be $0.102 \text{ dm}^3 \text{ mol}^{-1} \text{ s}^{-1}$ at $25.9 \text{ }^\circ\text{C}$. Typical data collected by the students are shown in Figure 2.

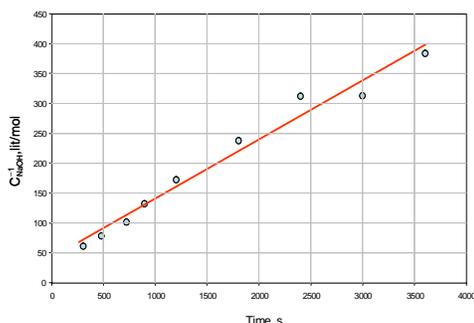


Fig. 2 Analysis of kinetic data obtained in a batch reactor

These results agree with those reported in the literature; the rate constant is reported to be equal to $0.111 \text{ dm}^3 \text{ mol}^{-1} \text{ s}^{-1}$ at $25 \text{ }^\circ\text{C}$ in Bamford and Tipper (1972).

The same reaction was carried out, at the same temperature, in the tubular reactor to investigate its performance in the laminar-flow range. The effect of Reynolds number on conversion of sodium hydroxide is shown in Figure 3.

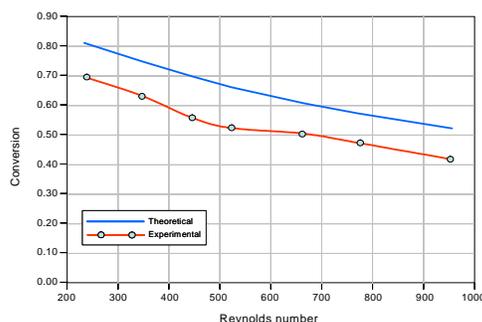


Fig. 3 Effect of Reynolds number in LFR

An inverse relationship between Re and conversion is observed: as Re increases the conversion at the outlet of the reactor decreases. The maximum discrepancy between the theoretical values of conversion, calculated using Eq. (4), and the experimental data in Figure 3 is 15 %. In our opinion such a discrepancy is acceptable in a demonstration experiment, considering errors inherent in collecting samples and in the analytical technique.

The dependence of the outlet conversion on space time (or mean residence time) is the focus of analysis in the CRE course, especially in the case of homogenous CSTRs and PFRs. Figure 4 nicely illustrates the effect of mean residence time on steady-state conversion in a LFR. As expected, conversion increases as space time increases. This relationship can be physically interpreted as the larger the space time, the smaller the volumetric flow rate and the better the conversion. The two curves in Figure 4 show a

maximum deviation around 20 % when the space time is 282 s. Again, we feel such a discrepancy is not uncommon in laboratory demonstrations.

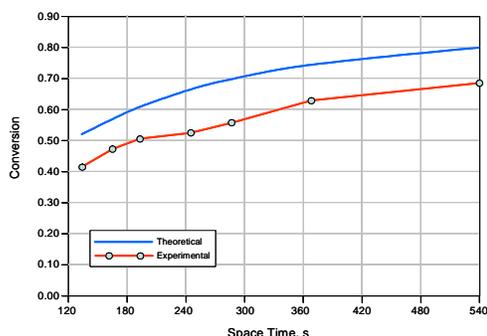


Fig. 4 Effect of space time on conversion in LFR

Concluding Remarks

A tubular reactor was operated isothermally in the laminar-flow regime to demonstrate basic transport phenomena concepts to students in the context of the undergraduate CRE laboratory. Students are able to test theories presented in class lectures: reaction kinetics; steady-state reactor performance; and space time versus real time. Concepts such as mixing-cup average concentrations, Reynolds number, and effect of tube bends can also be part of the discussion in this experiment.

The sodium hydroxide-ethyl acetate reaction system was chosen because of its well-known kinetics that our students confirm in batch-reactor experiments in the CRE laboratory. The relative ease of the analytical technique, which is based on direct sampling and titration, is another reason for this choice of the reaction system. Although the technique can be tedious for students, careful planning is necessary to ensure the experiments are completed within the duration of the laboratory session.

In the comparisons of theoretical and experimental results we see reasonable agreement. There might be other reasons to explain deviations,

such as the assumption of pure convection (Levenspiel, 1999) which is implicit in the use of Eq. (1) as a model of the reactor system. Indeed, there may well be some degree of molecular diffusion in the reactor. As this subject is not traditionally covered in our undergraduate reaction engineering course, we do not expect the students to discuss this issue in the analysis of the results.

At the end of our laboratory experiments on tubular reactors, we hope the difference between laminar- and plug-flow reactors is clearly demarcated.

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Nomenclature

C_A	concentration	[mol .dm ³]
C_{A0}	feed concentration	[mol .dm ³]
D	inside diameter of tube	[m]
k	reaction rate constant	
L	reactor length	[m]
r	radial position	[m]
$(-R_A)$	reaction rate	[mol.dm ⁻³ .s ⁻¹]
Re	Reynolds number, $\rho u_0 D / \mu$	
u	velocity	[m.s ⁻¹]
u_0	superficial velocity	[m.s ⁻¹]
V	reactor volume	[m ³]
v_0	volumetric flow rate	[m ³ .s ⁻¹]
x_A	conversion of reactant A	
z	axial position	[m]
ρ	density of mixture	[kg.m ⁻³]
μ	viscosity of reaction mixture	[Pa]
τ	space time, V/v_0	[s]

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