A Simple Experiment
FOR TEACHING PROCESS INTENSIFICATION BY STATIC MIXING
In Chemical Reaction Engineering

Sergio A. Baz-Rodríguez, Natali C. Herrera-Soberanis, Miguel Rodríguez-Novelo,
Juana A. Guillén-Francisco, and José A. Rocha-Uribe
Universidad Autónoma de Yucatán • Merida, Yuc., Mexico

The general contents of undergraduate courses in chemical reaction engineering (CRE) remain strongly influenced by the traditional conception outlined by prominent chemical engineering educators in the '50s and '60s of the past century. Even now, a typical reaction engineering course is centered on the theoretical analysis of ideal homogeneous reactors, and it is often supported by a set of related experimental practices in the laboratory. Certainly, the simplicity of ideal models is a great advantage when the students first approach the subject. However, it is important to be aware of the constraints of some concepts often employed in the courses, such as those pointed out by Churchill[1] in his interesting commentary on the state-of-the-art of education in reaction engineering. Also, and concurrent with the above, it is advantageous to enhance the chemical reaction engineering courses with relevant concepts of computational fluid dynamics and process intensification, areas that had great advances during the past two decades within reaction engineering.

Including topics of process intensification into reaction engineering is motivated by the inadequacy of traditional teaching schemes in reaction engineering, since they are developed to minimize the interdependence among transfer processes by means of simplifying assumptions. For example, as a consequence of ideal flow pattern assumptions (three- or two-dimensional perfect mixing for agitated and tubular reactors, respectively), the interrelation between momentum and mass transfer is reduced to the minimum. The same happens in separation processes courses, since they are traditionally conceptualized in terms of "unit processes" in which predominant mass transfer driving forces are individually studied. In reaction engineering, only the coupling between thermal energy and reactive mass balances is usually addressed.

Process intensification comprises the development of novel equipment and processes that offer substantial improvements (*e.g.*, smaller equipment and/or operation time, less energy consumption) with respect to conventional ones in chemical engineering.[2] In the framework of reactor engineering, reactive separations and intensified mixed reactors are the main approaches of process intensification. In both cases, hybrid

***Sergio A. Baz-Rodríguez** is a professor of chemical engineering at Universidad Autónoma de Yucatán, México. He received his B.Sc. at Universidad Autónoma de Yucatán (2005), his M.Sc. at Instituto Tecnológico de Celaya (2008), and his Ph.D. at Universidad Autónoma Metropolitana – Iztapalapa (2012), in México. His main field of research is transport phenomena in multiphase systems, particularly mathematical up-scaling of momentum and mass transfer in gas-liquid, solid-fluid, and porous media, CFD, and experimental mass transfer in bioreactors.*

***Natali C. Herrera-Soberanis**, **Miguel Rodríguez-Novelo**, and **Juana A. Guillén-Francisco** are undergraduate students in the Chemical Engineering Program of the Faculty of Chemical Engineering, at the Universidad Autónoma de Yucatán, México.*

***José A. Rocha-Uribe** is a professor of chemical engineering at Universidad Autónoma de Yucatán, México. He received his B.Sc. at Instituto Tecnológico de Celaya, México (1969), his M.Sc. at Instituto Politécnico Nacional, México (1977), and his Ph.D. at University of Texas at Austin (1984). His fields of research include separation processes by liquid-liquid extraction, distillation, and supercritical conditions.*

processes are involved: (i) simultaneous reaction and interfacial species transfer for reactive separations, and (ii) simultaneous reaction and momentum transfer management at different mixing scales for intensified mixed reactors.

The development and practical employment of hybrid processes in academic research and industry make it necessary to improve the teaching of chemical engineering in this area. Accordingly, simultaneous analysis of coupled transfer processes without minimizing the overall contribution of any of them is required. The main equipment for process intensification by enhanced mixing in reaction engineering includes microchannel reactors, spinning disk reactors, cavitation reactors, and static mixer reactors. In them, an enhanced mixing—and reaction—is achieved by passing the reactive flow through very thin conduits and/or moving walls, by the promotion of a spontaneous and turbulent change of phase, and by the inclusion of mechanical devices that constantly redistribute and mix a preferably axial flow, respectively.[3]

In this contribution, a simple experiment for static mixing in CRE is presented. A reaction test is carried out in a tubular reactor that acts as either a static mixer reactor or a laminar flow reactor; this experiment can be useful for teaching (i) the laminar flow reactor model, (ii) basic applications of macromixing intensification for enhancing the reaction extent, and (iii) models for pressure drop applied to static mixer devices.

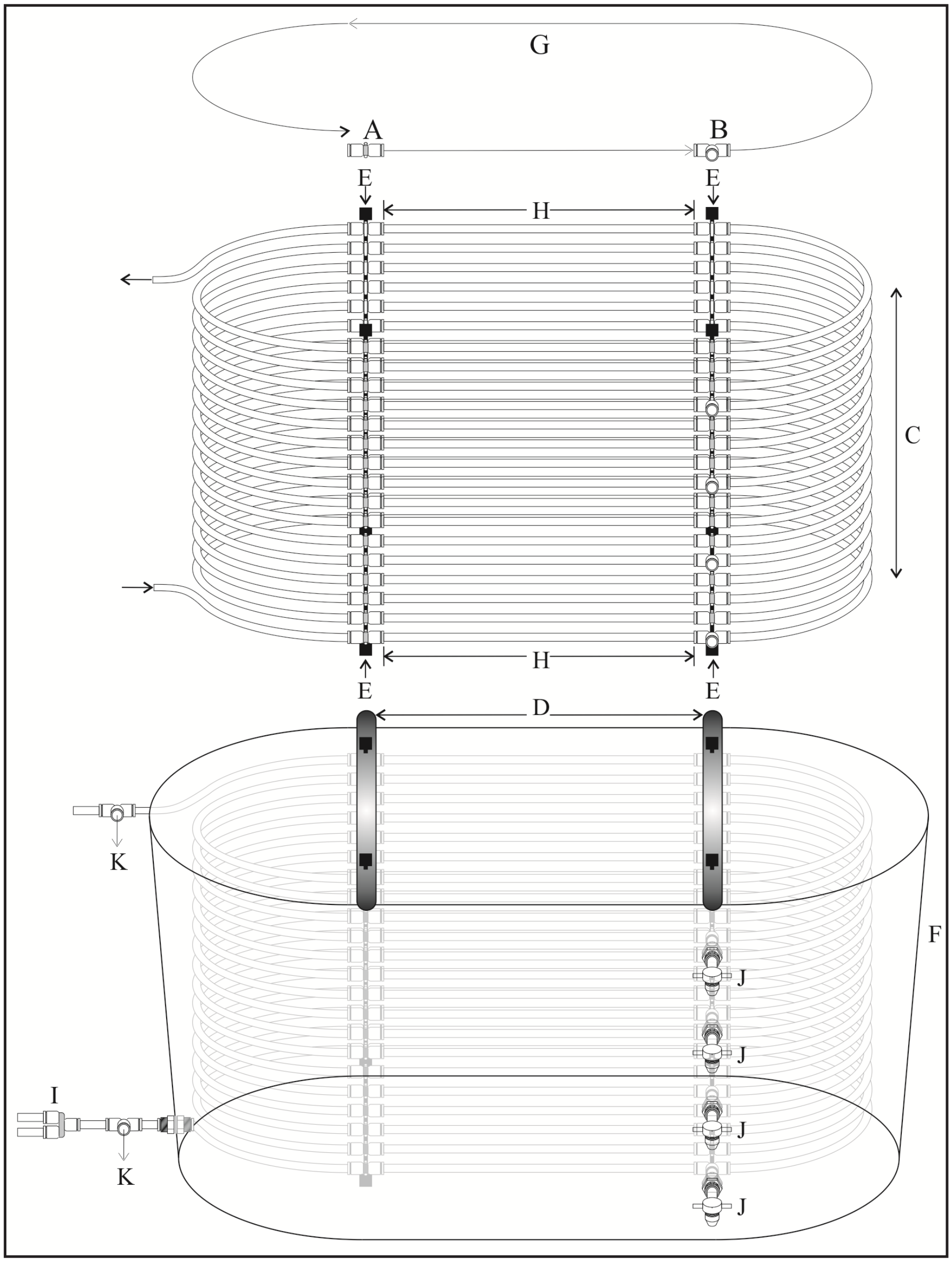

__Figure 1.__ Scheme of the designed tubular reactor.

APPARATUS

A simple static mixer reactor was designed and built. Helical static mixer elements were fabricated and inserted into straight sections of the reactor. The equipment acts as a laminar flow reactor when the static mixing elements are withdrawn. Figure 1 shows a scheme of the tubular reactor. The reactor is constructed with polyurethane hose sections ¼” NPT (TIUB07, SMC Pneumatics) connected by “push-in” straight unions (A) (C20200400, IMI Norgren) and tee unions (B) (C20600400, IMI Norgren, these for sampling points only); it is arranged as a coil with 16 turns (C), and the coil was hung in two aluminum rods (D) using four steel wires (E), and settled into an acrylic can (F). The total length of the reactor is 15.79 m, and its inner diameter is 0.00423 m. Into each turn of the coil (0.94 m long) (G), two parallel sections of 0.22 m with removable motionless mixers of 0.205 m long are placed (H). The feed of two different reactant solutions to the reactor is supplied employing two solenoid metering pumps (gamma/L, ProMinent Dosiertechnik GmbH) in another piece of equipment from our chemical engineering laboratory. The pump discharge lines are joined by a

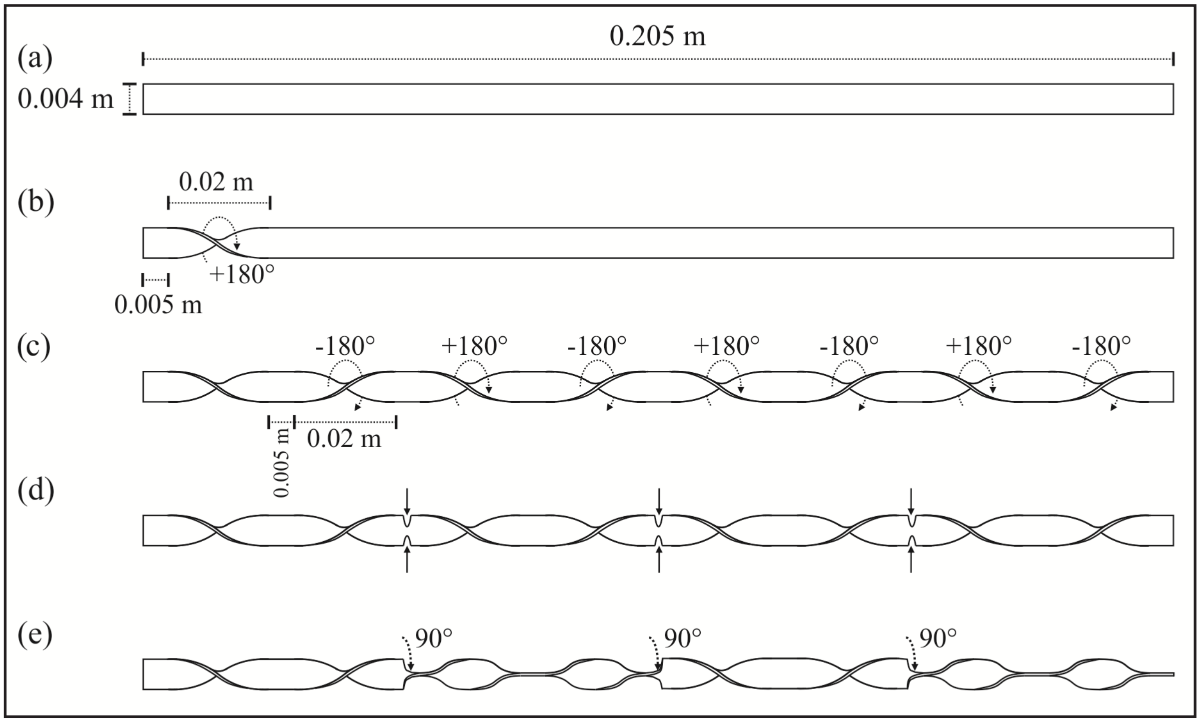

Figure 2. *Helical static mixer elements from stainless steel strips.*

"push-in" Y union (C20820400, IMI Norgren) and connected to the reactor inlet (I). In addition to the reactor outlet, four sampling lateral outlets are enabled (J); they pass through the can wall using "push-in" bulkhead unions (C20290400, IMI Norgren) and discharge liquid by opening aquarium globe valves; the sampling positions were at 0.945, 4.705, 8.465, and 12.225 m from the inlet. Also, for measuring pressure drop, the inlet and outlet of the reactor were laterally connected by tee unions to an available mercury manometer (K).

The elaboration of helical static mixer elements is schematized in Figure 2, and it was assembled as follows: From a sheet of stainless steel schedule 28 (0.000385 m thick), 32 strips were cut (0.004 m × 0.205 m) with an industrial shear (Figure 2a). In each strip, helical twists were made alternately in left- and right-hand directions with a turn angle of 180° in the following way: initially, 0.005 m from the left edge to 0.02 m ahead, a first twist was made fixing the extreme of strip and twisting with a pincer to give a clockwise rotation (Figure 2b); next, 0.005 m forward from the end of the first twist, and up to 0.02 m ahead, a second twist was made but with a counterclockwise direction. This procedure was repeated to give a series of twists in opposite directions, each pair being spaced 0.005 m between them (Figure 2c). Then, a pair of narrow notches was made with a lathe in the space between twists after each two adjacent twists (Figure 2d). Finally, in the positions where each pair of notches was made, the trailing edge of the twist forward was bent by hand to form and angle of 90° with the leading edge of the preceding twist (Figure 2e). The resulting 32 stainless steel strips configured as helical static mixers were introduced inside the hose sections disposed for mixing (sections H in Figure 1).

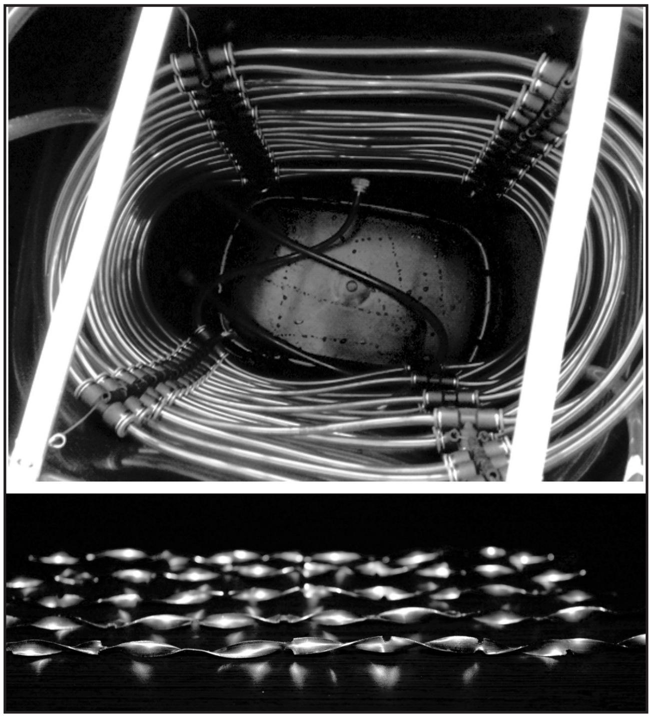

Figure 3. *Views of the reactor and the mixing elements.*

Pictures of the upper view of the reactor and the mixing elements are showed in Figure 3. Since the pumps and the manometer were available from another piece of equipment in our laboratory (both are usual elements in chemical engineering laboratories), the total cost of materials and construction of this simple equipment was less than $200 USD.

EXPERIMENTAL PROCEDURE

The reaction of saponification of ethyl acetate is often employed for supporting experimental teaching of chemical kinetics and reaction engineering at the undergraduate level[4-6] and it was chosen for testing the effects of static mixing on the reaction extent. The reaction under consideration is as follows:

$$CH_3COOCH_2CH_3 + NaOH \rightarrow CH_3COONa + CH_2CH_3OH \qquad (1)$$

In general, monitoring progress of this reaction is carried out by employing a conductivity electrode. However, since the thin inner diameter of the reactor hinders invasive monitoring with conventional electrodes, an alternative titration-based procedure was carried out for the experiments.[5,7-9]

As a reference for the operation of the reactor, a fixed Reynolds number (Re) around 350 (referred to the inner diameter of the reactor and the properties of water at 35 °C) was used. Thus, the flow regime for the reactor without the motionless mixer elements must be laminar. This Re implies a spatial time of around 265 s, which helps make the time for sampling and quenching the reaction (the samples are withdrawn directly from the reactor) significantly smaller than the characteristic global time of operation (according to the reported kinetics, at 35 °C the significant changes occur before 10-15 minutes). A high Re (close to turbulent transition) could make the sampling time of similar order of magnitude to the spatial time and it also could confine the concentration changes to the initial section of the reactor; on the other hand, a small Re (close to creeping flow) could damp the mixing effects of static mixers and make the concentration changes along the reactor only slightly perceptible.

The experimental procedure is summarized as follows:

1. The outlet of each pump is connected to the Y union at the inlet of the static mixing reactor; for measuring pressure drop, two lateral outlets are enabled by tee unions at the inlet and outlet of the reactor, respectively, and connected to a mercury manometer.
2. 1.5 liters of two solutions 0.1 M of ethyl acetate and sodium hydroxide, respectively, are prepared and put into the feed tank of each pump.
3. The pumps are calibrated and a volumetric flow of 25 mL/min is specified for feeding each reactant solution; thus, a total flow (Q) of 50 mL/min 0.05 M of each reactant enters the reactor ($c_{CH_3COOCH_2CH_3,0} = c_{NaOH,0} = 0.05$ M).
4. Since the space time is around 4.43 min, the reactive mixture is allowed to flow through the reactor for about 15 min.
5. At the outlet of the reactor, a sample of 5 mL is withdrawn into a test tube containing 5 mL of a quenching solution of 0.05 M HCl. Immediately, the partially mixed sample in quenching is added to a flask with 5 mL of water being magnetically agitated in order to complete the interruption of the reaction. The quenching solution is prepared in such a way that 5 mL of it has HCl in excess with respect to any possible quantity of NaOH contained in the sample.
6. The sample in the flask is titrated with a solution 0.025 M NaOH using blue bromothymol as indicator. The concentration of NaOH in the original sample is then calculated from stoichiometry and material balances.
7. Steps 5 and 6 are repeated but now we withdraw the samples in each one of the lateral outlets, beginning at the nearest to the outlet section and finishing at the nearest to the inlet of the reactor. The above is in order to avoid disturbing the flow distribution in the remaining non-sampled points of the equipment. The withdrawn samples are carried out by plugging up the outlet of the reactor and simultaneously opening the valve of each lateral point of sampling, and after draining the stagnant liquid between the valve and the main body of the reactor.
8. The procedure (steps 1 to 7) is repeated but with the reactor operating without the static mixer elements, in order to evaluate and match the effects of motionless mixers with respect to the performance under a laminar flow regime.

DATA ANALYSIS

Students grouped by teams carried out the reaction tests during their course of Reactor Engineering and a Research Workshop. Figure 4 shows the obtained average

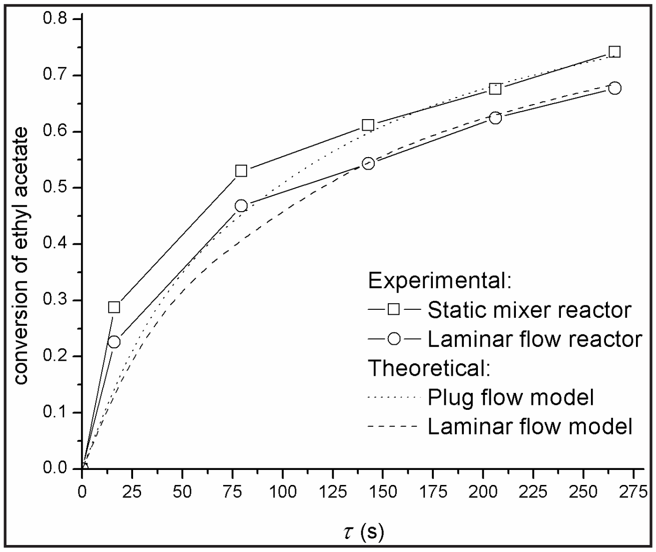

Figure 4. *Profiles of conversion of ethyl acetate in the tubular reactor at 35 °C.*

results for reactant conversion. Larger conversions of reactant were achieved when motionless mixer elements were included along the tubular reactor. At the outlet, fractional conversions of 0.74 for the static mixer configuration and 0.68 for the laminar flow configuration were measured, respectively.

The calculation of theoretical conversion profiles and their matching with experimental results are essential to reinforce the understanding of models and to give the students a view of the scope of intensified systems. Thus, for a best assessment of the results, it is useful to encourage the students to compare results with two theoretical models: the classical plug flow and the laminar flow models. The first one is a well-known element for reaction engineering courses; the second one is usually tackled in the framework of residence time distributions (RTD) and the segregated flow model for designing continuous reactors.[11,12] The reaction test proposed here provides support for introducing and comparing theory and experiments. Since the reaction is just slightly exothermic, the thermal effects can be neglected.

Eqs. (2)–(5) are employed to calculate theoretical conversions (X).

Reaction rate[10]:

$$-r_{CH_3COOCH_2CH_3} = kc_{CH_3COOCH_2CH_3}c_{NaOH} \tag{2}$$

$$k = 28.57\times10^6\left(\frac{L}{mol\cdot s}\right)\exp\left(\frac{-48000\dfrac{J}{mol}}{RT}\right) \tag{3}$$

Laminar flow reactor (2nd order kinetics)[11] :

$$X_{CH_3COOCH_2CH_3} = kc_{CH_3COOCH_2CH_3,0}\tau\left[1-\frac{kc_{CH_3COOCH_2CH_3,0}\tau}{2}\ln\left(1+\frac{2}{kc_{CH_3COOCH_2CH_3,0}\tau}\right)\right] \tag{4}$$

Plug flow reactor (2nd order kinetics):

$$X_{CH_3COOCH_2CH_3} = \frac{k\tau c_{CH_3COOCH_2CH_3,0}}{1+k\tau c_{CH_3COOCH_2CH_3,0}} \tag{5}$$

From the second order kinetics, the space time ($\tau = 265$ s) and the local average temperature in the range of time when the experiments were made in Mérida, México (T = 35 °C, second and first fortnight of March and April, 2015, respectively), the theoretical conversion profiles were calculated, and they are also reported in Figure 4.

At the outlet, fractional conversions of 0.73 for the plug flow model and 0.69 for the laminar flow model were reported. Apparently, an excellent agreement between theory and experiments is achieved for the laminar flow operation, and the motionless mixing operation approaches the plug flow theory. However, the conversion data for the first three sampling points, which are larger than theoretical maxima, suggest a delay effect due to the sampling times. It is likely that in the time that elapses from the sampling to the total quenching, the reaction continues occurring. In the first sampling points, where there are larger concentrations of reactants and, consequently, higher reaction rates could be expected, the sampling and quenching times probably allow that the reaction significantly continues by some additional seconds. Thus, higher conversions than theoretical predictions are measured. A feasible solution for this issue would be to incorporate miniature flow through conductivity electrodes at the sampling points and change the reaction monitoring method (in-line measurement), but an additional and comparatively significant investment would be required. Beyond the above, the trends of experimental conversion profiles showed in Figure 4 indicate the reaction intensification due to the static mixer elements.

The behavior tending to a plug flow model when the static mixer elements are inserted into the tubular reactor is related to the degree of cross-sectional concentration homogeneity that they promote. Although out of the scope of the data results of this experiment, the students would be encouraged to understand the quantitative description of the mixture quality in terms, for example, of coefficients of variation—which are the ratio of the standard deviation in tracer composition to the mean composition in cross-sections along the reactor.[13]

The characterization of non-ideal flow patterns by RTD is generally included in the goals of a CRE course. Then, an overview of the known behavior of RTD for tubular conduits with helical static mixers[14,15] could also help to clarify for students the hydrodynamic effects into the reactor. The peak of the curve of RTD increases and the curve form narrows as more elements are inserted, being an indication of more uniformity for the radial concentration around a plug element. The experimental verification from RTD can be carried out from a pulse injection of concentrated solution of a salt through a water feed at the inlet, followed by the monitoring of the transient concentration at the outlet.

The mixing intensification due to the motionless mixer elements occurs at the expense of the increment of the pressure drop along the reactor ($-\Delta P$), and this must be clear for the students. The feasibility of a static mixer reactor depends to a good extent on the additional energy consumption because of the increments of pressure drop by the static mixing and the relative enhancement in the reaction performance.

The experimental results for pressure drop when the equipment operates as a laminar flow reactor can be compared with those calculated by the Fanning Equation with a laminar friction factor—linking reaction engineering with traditional concepts of applied momentum transfer courses:

$$-\Delta P = \frac{32 f \rho Q^2 L}{\pi^2 d_i^5} \tag{6}$$

$$f = \frac{16}{Re} \tag{7}$$

where ρ is the fluid density, L is the total length of the reactor, d_i is the inner diameter and f is the Fanning friction factor. Both the results of experimental tests and the theory for the laminar case are reported in Table 1. A moderate agreement (absolute relative error around of 22 %) is observed; the differences can be discussed with students in terms of the neglected effects of coiling. If it is of teaching interest, friction factor correlations for helical coil tubes (f_c) can be evaluated and used in the Fanning Equation for pressure drop calculation. Two suggestions are the correlations for laminar regime of Srinivasan, et al.[16] and Manlapaz and Churchill.[17] An increment around 15-25 % with respect to the straight pipe pressure drop is calculated this way, giving predictions closer to the experiments. Additional parameters such as the ratio between the hose inner diameter (d_i) and the equivalent diameter of the hose coil (D_{eq}), and the Dean number ($De = Re\sqrt{d_i/D_{eq}}$), are relevant for calculation of pressure drop in the coil (for the constructed tubular reactor: $D_{eq} \approx 0.29$ m; De = 42.3).

TABLE 1
Pressure drop in the tubular reactor

		-ΔP (Pa)
Laminar flow reactor	Experiment	1473
	Theory	1206
Static mixer reactor	Experiment	4914
	Correlation (Song, et al.[19])	4794

The measured pressure drop for the static mixer configuration can be compared to pertinent correlations for pipes with motionless mixers. The elements employed in this contribution have similitudes with the well-known Kenics helical elements (KMS, Chemineer, Inc.). The motionless elements of this work differ from the geometry of Kenics elements since (i) the bendings of 90° between edge elements are made after each two alternative twists and not after one, (ii) the ratio of diameter to length of a twist in the mixing element (TR) is higher (5 vs. 1.5), and (iii) there is a space (0.005 m) between twists. However, for comparison purposes, the available correlations in the literature for pressure drop with helical elements type Kenics can be useful. From dimensionless analysis it has been identified that the friction factor mainly depends on Re (referred to the empty pipe) and TR.[18,19] From Song, et al.,[19] the Fanning friction factor can be expressed as follows:

$$f = \frac{K\left(Re/TR^{2.15}\right)^n}{4TR^{2.04}} \tag{8}$$

with K = 320 and n = -0.86 for the approximated geometry and Re employed in this experiment. The total pressure drop can be obtained as follows:

$$-\Delta P = \left(-\Delta P_{SM}\right) + \left(-\Delta P_{LF}\right) \tag{9}$$

with $-\Delta P_{SM}$ as the pressure drop in the static mixing sections and $-\Delta P_{LF}$ as the pressure drop in the laminar flow sections. The Fanning equation can be used to calculate $-\Delta P_{SM}$ and $-\Delta P_{LF}$, but for the former Eq. (8) must be employed to calculate the friction factor. Also, the total length for the static mixing sections is 6.4 m and for the laminar flow sections is 9.39 m. Results of both the experimental test and the correlation for static mixing are also reported in Table 1. A good agreement (absolute relative error around of 2.5 %) is observed between them. If it is of interest, the effects of coiling can be taken into account here.

ASSESSMENT OF THE EXPERIMENT

Along with complementary experiments for single and in series CSTRs, the experiment described in this paper has been assigned as part of a final set of lab practices to students of the course Reactor Engineering (3rd year) and a Research Workshop (4th year) at the Universidad Autónoma de Yucatán, in 2015 (except CSTRs in series) and 2016. By ensuring that all the systems (static mixer reactor, laminar flow reactor, single CSTR, and CSTRs in series) operate with the same reaction and global space time, the students have a similar basis for comparison for the full experimental assessment of isothermal homogeneous reactors. Moreover, regarding the analysis of the static mixer reactor, additional issues related to mixing intensification, pressure drop measurement, and calculation were covered.

Traditional courses of CRE often emphasize the analysis of heat requirements for ideal reactor models. However, more emphasis must be put on power requirements for fluid flow in reactors, either for overcoming pressure drop due to friction or for providing mechanical agitation. The conceptual design of homogeneous reactors must take into account the reaction extent together with heat and power requirements. In our experience, the proposed experiment helps students to clearly understand that adding internals to tubular reactors would enhance the reaction extent but has a price in terms of the extra pressure drop. Also, the proposed experiment reinforces the connection between basic concepts of applied momentum transfer courses (friction factor and pressure drop calculations) and reaction engineering. Finally, like many laboratory experiments in chemical engineering, matching between theory and experiments is fundamental for emphasizing the relevance and usefulness of theory and correlations that are covered in the classroom.

The scope of this experiment is focused and limited to evaluating general macromixing effects under a reaction-based point of view. For a complementary analysis in the framework of hydrodynamics, the axial dispersion and predictions of reaction extent by the segregated flow model can be easily obtained from an RTD.

CONCLUSIONS

Process intensification is a trend in chemical engineering, and the proposed experiment provides material support for teaching mixing intensification in a course of CRE. Since the experimental results allow viewing the effect of static mixing both in the reaction conversion and in the pressure drop, this proposal can be useful for introducing topics of macromixing intensification and for reinforcing essential concepts of homogeneous reactor design and pressure drop calculations.

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REFERENCES

1. Churchill S.W., "Commentary: The State of the Art of Education in Reaction Engineering," *Ind. Eng. Chem. Res.*, **50**, 8806 (2011)
2. Stankiewicz, A., and J.A. Moulijn, "Process Intensification: Transforming Chemical Engineering," *Chem. Eng. Progress*, January: 22-43 (2000)
3. Stankiewicz, A., and J.A. Moulijn, *Re-Engineering the Chemical Process Plant: Process Intensification*, Marcel Dekker, Inc., US (2004)
4. Lindfords, L.-E., "An Undergraduate Experiment in Chemical Engineering Reactor Kinetics," *J. Chem. Educ.*, **48**, 472 (1971)
5. Sundberg, D.C., T.E. Carleson, and R.D. McCollister, "Experiments in Undergraduate Reaction Engineering: Startup and Transient Response of CSTR's in Series," *Chem. Eng. Ed.*, **15**(3), 118 (1977)
6. Mendes, A.M., L.M. Madeira, F.D. Magalhaes, and J.M. Sousa, "An Integrated Chemical Reaction Engineering Lab Experiment," *Chem. Eng. Ed.*, **38**(3), 228 (2004)
7. Smith, H., "Kinetics of the Catalyzed Esterification of Normal Aliphatic Acids in Methyl Alcohol," *J. Am. Chem. Soc.*, **61**, 254 (1935)
8. Evans, D.P., J.J. Gordon, and H.B. Watson, "Studies of the Ortho-Effect. Part III. Alkaline Hydrolysis of Benzoic Esters," *J. Chem. Soc.*, 1430 (1937)
9. Potts, J.E., and E.S. Amis, "The Alkaline Hydrolysis of Ethyl Acetate From the Standpoint of Ion-Dipole Theory," *J. Am. Chem. Soc.*, **71**, 2112 (1949)
10. Richter, O., M. Menges, and B. Kraushaar-Czarnetzki, "Investigation of Mixing In A Rotor Shape Modified Taylor-Vortex Reactor By the Means of a Chemical Test Reaction," *Chem. Eng. Sci.*, **64**, 2384 (2009)
11. Levenspiel, O., *Chemical Reaction Engineering*, 3rd Ed., John Wiley & Sons, US (1999)
12. Tiscareño-Lechuga, F., *ABC para comprender Reactores Químicos con Multireacción*, Instituto Tecnológico de Celaya - Editorial Reverté, México (2008)
13. Hobbs, D.M., and F.J. Mucio, "The Kenics Static Mixer: A Three-Dimensional Chaotic Flow," *Chem. Eng. J.*, **67**, 153 (1997)
14. Pustelnik, P., "Investigation of Residence Time Distribution In Kenics Static Mixers," *Chem. Eng. Processing*, **20**, 147 (1986)
15. Jin, D., H. Fu, J. Wu, and D. Sun, "Quantitative Analysis of Residence Time Distribution In Kenics Static Mixer," *Adv. Mat. Res.*, **499**, 198 (2012)
16. Srinivasan, P.S., S.S. Nandapurkar, and F.A. Holland, "Friction For Coils," *Trans. IChEME*, **48**, T156 (1980)
17. Manlapaz, R.L., and S.W. Churchill, "Fully Developed Laminar Flow In a Helically Coiled Tube of Finite Pinch," *Chem. Eng. Commun.*, **7**, 557 (1980).
18. Lecjaks, Z., I. Machac, and J. Sir, "Pressure Loss In Fluids Flowing In Pipes Equipped With Helical Screws," *Int. Chem. Eng.*, **27**, 205 (1987)
19. Song, H.-S., and S.P. Han, "A General Correlation For Pressure Drop In A Kenics Static Mixer," *Chem. Eng. Sci.*, **60**, 5696 (2005) ❒