

연속식 탱크 반응기에서의 혼합효과

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Mixing Effects in a Stirred Tank Reactor

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요 약

반응기 입구농도에 계단 입력을 주었을 때 과도응답을 측정함으로써 연속식 탱크 반응기의 비 이상 혼합효과를 조사하였다. 탱크의 레이놀즈수가 17을 넘으면 들어가는 유량의 난류만으로 완전혼합을 얻을 수 있었고 17 이하에서는 임펠라의 직경을 기준으로 한 레이놀즈수의 최소치를 탱크의 레이놀즈수와 관련시켜 $Re_m = 540 \ln(17/Re_t)$ 의 관계식을 얻었다. 입구관과 출구관을 넘쳐 흐르는 수위와 일치선으로 했을 때 by passing 효과를 조사했고 비 이상 흐름에서는 여러가지의 mixed model 로써 흐름의 pattern 을 나타냈다.

Abstract

Nonideal behaviour of a continuous flow stirred tank reactor was investigated by observing the transient response of the reactor to step changes in feed concentration. It was shown that the turbulence caused by inlet flow only was sufficient for perfect mixing provided that the tank Reynolds number exceeded 17. For the tank Reynolds number below 17, mechanical stirring was necessary and the Reynolds number based on the impeller diameter for perfect mixing was correlated to the tank Reynolds number as follows:

$$Re_m = 540 \ln(17/Re_t)$$

It was also found that bypassing effects became more significant when the feedline and effluent line were placed near the liquid levels. For nonideal flows, several mixed models described the flow patterns, namely bypassing and dead space model, CSTR and PFR in series model, and dead space model.

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Introduction

For the prediction of the performance of a flow reactor, two types of information are generally required, namely flow pattern and the information on the rate phenomena. If an ideal case of complete mixing or perfect piston flow can be taken to describe the flow behaviour with sufficient accuracy, the problem involving the flow pattern becomes simple. In other cases it is possible by various methods to establish experimentally the distribution of residence times (RTD) of the flowing material.

It was Danckwerts¹⁾ who first introduced the concept of residence time distribution. He made it clear that it is essential to distinguish between "macromixing" and "micromixing" phenomena, and that RTD studies can contribute only to a better understanding of the former. For a first-order reaction, the rates are not affected by the interaction between fluid elements and the reactor performance can be described by the macromixing information alone. For the other kinetics, it is necessary to obtain micromixing information for the complete description of the flow behaviour. Zwietering²⁾ determined the two bounds for micromixing theoretically, namely "maximum mixedness" and "complete segregation". These bounds are often sufficiently narrow³⁾ and the flow patterns of most industrial reactor can be described by the macromixing information alone for all practicality.

RTD curves for continuous flow stirred tank reactors (CFSTR) can be experimentally obtained by observing the transient response to a step input of a tracer material. Such an attempt was first made by Cholette and Cloutier⁴⁾ who obtained RTD curves at different stirring rates with a conveniently designed CFSTR for nonideal mixing. Other investigators reported RTD information for different physical parameter such as fluid viscosity, flow rate, tank geometry, impeller diameter, etc.^{5,6)}

In the present investigation, the region of ideal mixing without mechanical stirring is first determined. For the nonideal mixing region, RTD curves at different tank Reynolds numbers and impeller Reynolds

numbers are obtained experimentally and their results are correlated in an attempt for the broader knowledge on the mixing phenomena in a CFSTR.

Theory

Mixing in a CFSTR is caused by the following two factors: (1) the kinetic energy transported to the tank with the inlet flow; and (2) the mechanical stirring by the impeller. Most of the actual processes require the installation of impellers, because the process flow rates are usually insufficient for the desired level of mixing. Therefore, it is necessary to define separately the degree of turbulence caused by the flow and that caused by the impeller. The former can be expressed by the modified Reynolds number based on the tank diameter, Re_t ,

$$Re_t = \frac{D_i G}{\mu} \quad \text{where } G = \frac{Q_0}{\pi D_i^2 / 4} \quad (1)$$

where G denotes the mass velocity. Similarly, another modified Reynolds number for the latter may be defined as follows:

$$Re_m = \frac{N d_m^2 \rho}{\mu} \quad (2)$$

The minimum value of Re_t for perfect mixing, Re_t^* , was experimentally determined. The design of a CFSTR with Re_t above Re_t^* becomes trivial because the ideal mixing will be achieved in such a case. In case when Re_t is smaller than Re_t^* , it is necessary to find the minimum value of Re_m for perfect mixing. An empirical correlation between Re_m^* and Re_t will be useful for the preliminary design of a CFSTR:

$$Re_m^* = f(Re_t^*/Re_t) \quad (3)$$

A logical procedure for the design of an imperfectly mixed CFSTR is as follows;

- (1) Determination of the residence time distribution with the tracer experiments without reaction;
- (2) Selection of a suitable mixed model that best describes the flow pattern shown by the residence time distribution;
- (3) Prediction of the performance of the reactor with reaction by the chosen mixed model.

The residence time distribution functions obtained from the tracer experiments were compared with those of three of the *a priori* mixed models. The internal

residence time distribution function, $I(\theta)$, is defined as $I(\theta) = \frac{C - C_F}{C_0 - C_F}$. The theoretical tracer curves for these models are listed below:⁷⁾

1) Dead Region and Bypassing Model

$$\ln \frac{C - C_F}{C_0 - C_F} = \ln \frac{Qa}{Q} - \frac{Qa}{Q} \cdot \frac{V}{Va} \cdot \theta \quad (4)$$

2) CFSTR and PFR in series Model

$$\ln \frac{C - C_F}{C_0 - C_F} = -\frac{V}{Va} \theta + \frac{V}{Va} \left(1 - \frac{Va}{V}\right) \quad (5)$$

3) Partial mixing with Piston Flow and Short Circuit

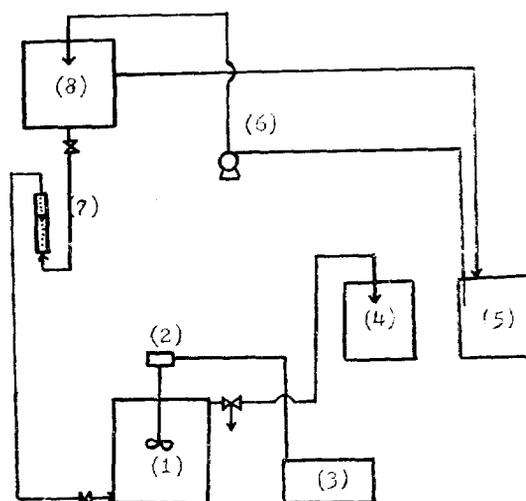
$$\frac{C - C_F}{C_0 - C_F} = \left[n + p \cdot \exp\left(\frac{(p+n)(1-m)}{mp}\right) \right] \exp\left[\frac{p+n}{m} \cdot \theta\right] \quad (6)$$

Experimental

Figure 1 shows the schematic diagram for the experimental system. The reactor used was an overflow reactor of height 18.3cm and internal diameter 15cm. The material for the reactor was stainless steel. Initially the tank was filled with 0.1N HCl solution. A four-blade impeller of diameter 3.5 cm was used for stirring. After running the agitator for some time at the desired speed to allow for the mixing pattern to be fully developed, water or glycerine solution was suddenly introduced through the constant head tank to the reactor at the required flow rates. Samples were drawn from the reactor effluent at regular time intervals for pH measurements. The pH measurements were made by a Fisher model 320 expanded scale pH meter. A rotameter was used for the measurement of flow rates that were regulated by a needle valve inserted in the feed line. The feed and exit lines were $\frac{1}{4}$ -in. copper tubing. The stirring rate for the impeller was regulated by the variable-speed motor and voltage transformer, and measured by a rpm tachometer.

The experiments were carried out with the two different feed positions.

In the first set of experiments the feed line was placed at the bottom of the reactor and the impeller position was at 8 cm from the bottom of the reactor. In the second set of experiments the feed line was moved to 1cm from the top of the reactor and the impeller to 3cm from the top.



(1) CSTR, (2) variable speed electrical motor, (3) voltage regulator, (4) outlet stream storage, (5) feed tank, (6) pump, (7) rotameter, (8) constant head tank.

Fig. 1. Schematic diagram of the RTD experiments.

Results and discussion

Table 1 shows the summary of the experimental results. Figures 2 through 8 are the residence time distribution functions for the various experimental conditions. Figure 2 shows that perfect mixing is obtained at Re_t greater than 17 without mechanical stirring when water is used as the flowing material. From this result, it may be concluded that little mechanical agitation is required for perfect mixing when the fluids with low viscosities are used for the flowing materials. As we increase the viscosities of the flowing material, more mechanical agitation will be necessary for complete mixing. For the fluids of higher viscosities, perfect mixing is attained if the tank Reynolds number exceeds 17 without mechanical stirring as shown in Fig. 3. Figure 4 shows RTD for perfect mixing attained at Re_t smaller than 17 with mechanical stirring. From the results of Fig. 4, 5, and 6, the minimum value of Re_m^* for perfect mixing is related to the tank Reynolds number as follows:

$$Re_m^* = 540 \ln(17/Re_t) \quad (7)$$

Table 1. Summary of the experimental results.

Run	Q (ml/min)	ρ (g/cm ³)	τ (min)	μ (cp)	N (rpm)	Re _t	Re _m	Pattern of flow
1	543	1.	5.76	1.	0	76.9	0	ideal
2	297	1.	10.53	1.	0	42.	0	"
3	155	1.	20.2	1.	0	21.9	0	"
4	120	1.	26.1	1.	0	16.9	0	Bypass and dead region
5	87	1.	36.	1.	0	12.3	0	nonideal
6	474	1.1	6.6	4.3	0	17.2	0	ideal
7	324	1.1	9.66	4.3	0	11.7	0	Bypass and dead region
8	255	1.1	12.3	4.3	9	9.25	0	nonideal
9	180	1.1	17.4	4.3	100	6.51	507	Bypass and dead region
10	330	1.1	9.5	4.3	122	12	619	ideal
11	330	1.1	9.5	4.3	140	12	710	ideal
12	417	1.1	7.5	7.2	0	9.25	0	Bypass and dead region
13	410	1.13	7.65	7.2	120	9.1	374	CSTR and PFR in series
14	420	1.13	7.45	7.2	164	9.32	511	ideal
15	330	1.13	9.5	7.2	164	7.32	511	ideal
16	260	1.13	12	7.2	164	5.77	511	CSTR and PFR in series
17	220	1.13	14.2	6.85	98	5.14	321	Bypass and dead region
18	230	1.13	13.6	6.85	48	5.37	154	CSTR and PFR in series
19	230	1.13	13.6	6.85	192	5.37	780	ideal
20	260	1.13	12.	6.85	200	5.77	750	ideal
21	325	1.17	9.65	16.7	0	3.22	0	nonideal
22	335	1.17	9.35	15.9	112	3.48	163.5	CSTR and PFR in series
23	318	1.17	9.85	15.9	180	3.3	262	"
24	292	1.17	10.06	17.46	80	2.8	106	"
25	315	1.17	9.95	16.2	130	3.22	186	"
26	230	1.17	13.6	18.5	86	2.06	108	"
27	335	1.17	9.35	15.9	623	3.48	910	ideal
28	315	1.17	9.95	16.2	510	3.22	730	nonideal
41	475	1.	6.6	1.	0	67.2		Bypass and dead region
42	70	1.	44.8	1.	0	9.9		"
43	555	1.	5.65	1.	0	78.5		"
44	145	1.	21.6	1.	0	20.5		"
45	460	1.	6.8	1.	254	65.		Dead region
46	468	1.	6.8	1.	150	65.		Bypass and dead region
47	210	1.	14.9	1.	150	29.7		nonideal
48	210	1.	14.9	1.	1590	29.7		Dead region
49	310	1.17	9.85	15.54	0	3.49		nonideal
50	330	1.17	9.85	15.54	138	3.52		Bypass and dead region

Note: The feed position for Run No. 1 through 28 is at the bottom of the tank.

The feed position for Run No. 41 through 50 is 1cm from the top.

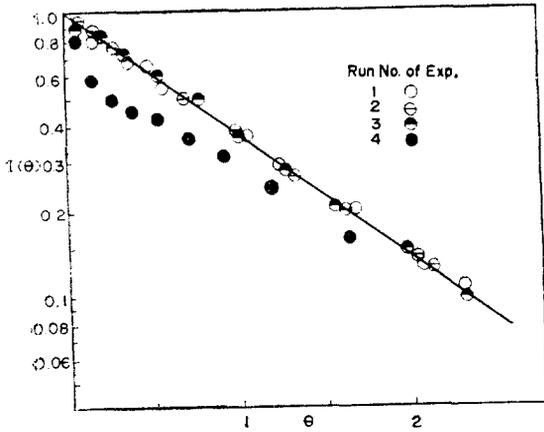


Fig. 2. Residence time distribution function $I(\theta)$ for viscosity $\mu=1\text{cp}$.

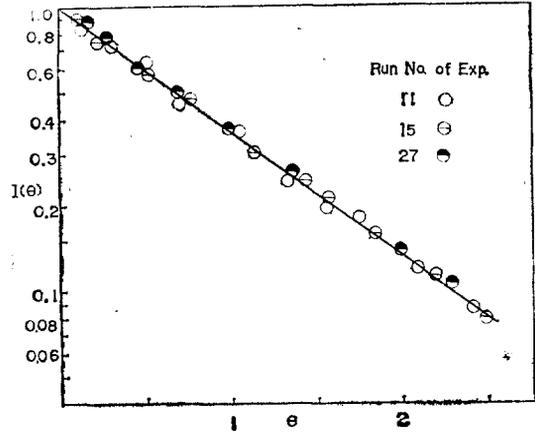


Fig. 4. RTD function $I(\theta)$ in the region $Re_t > 17$ and $Re_m > 540 \ln \frac{17}{Re_t}$.

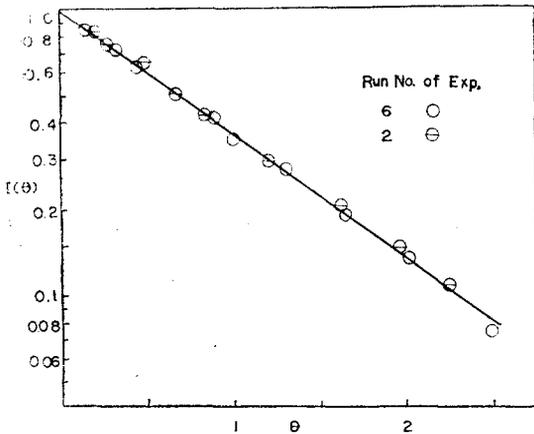


Fig. 3. RTD function $I(\theta)$ in the region of ideally mixed flow without mechanical mixing ($Re_t > 17$).

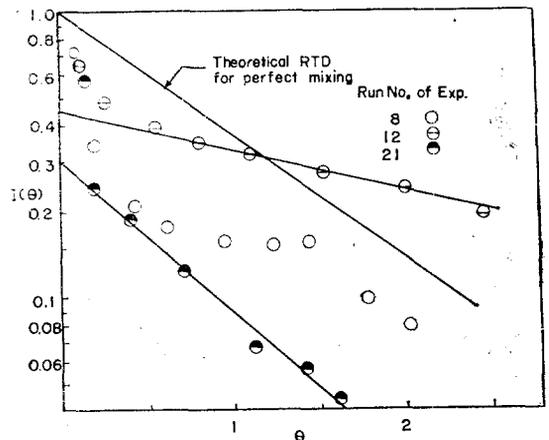


Fig. 5. RTD function $I(\theta)$ in the region of $Re_t < 17$ and $Re_m < 540 \ln \frac{17}{Re_t}$.

The results obtained by other investigators^{5,6} confirmed the validity of the ideally mixed regions described by the above equation.

Figures 5 through 8 are the residence time distribution functions for the case of nonideal mixing. As shown by the intercepts in Fig. 5 severe bypassing effects were shown when no mechanical agitation was applied. As we increase the stirring rate of the impeller, the intercept approaches unity, i.e., ideal mixing

state is approached, as shown in Fig. 6. Comparing these results with the residence time distribution functions derived in the theory section, it may be stated qualitatively that the bypass stream decreased with the increase of the tank Reynolds number. Figure 9 shows the plot of Re_t^*/Re_t vs. Re_m for various experiments. As shown in the figure, ideal and nonideal mixing regions are clearly divided by the line for Eq. (7).

Comparing the experimental results with the thoret-

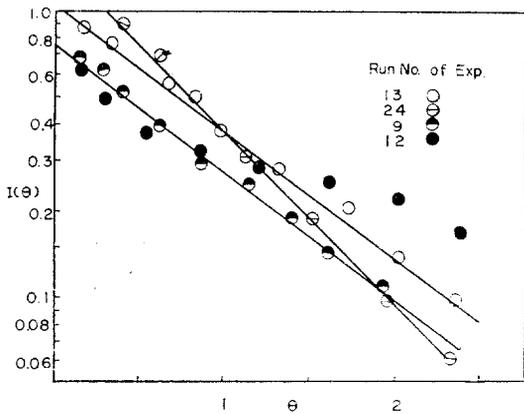


Fig. 6. RTD function $I(\theta)$ in the region of $Re_t < 17$ and $Re_m = 540 \ln \frac{17}{Re_t}$.

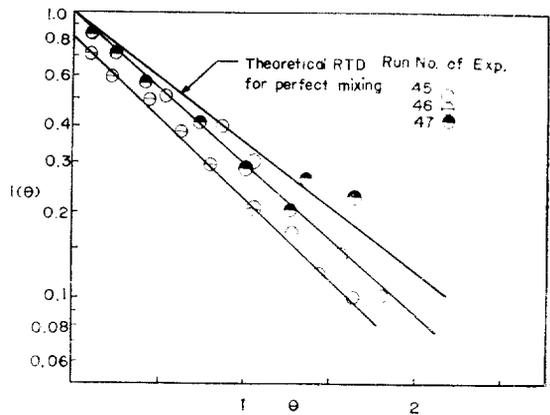


Fig. 8. RTD function $I(\theta)$ when the optimal geometry of the reactors are not preserved and impeller is used.

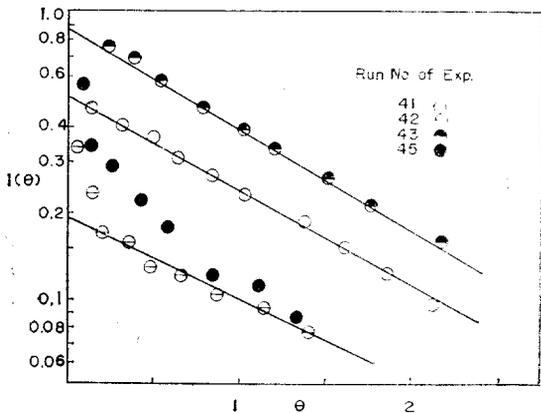


Fig. 7. RTD function $I(\theta)$ for viscosities $\mu=1cp$ when the optimal geometry of the reactors are not preserved.

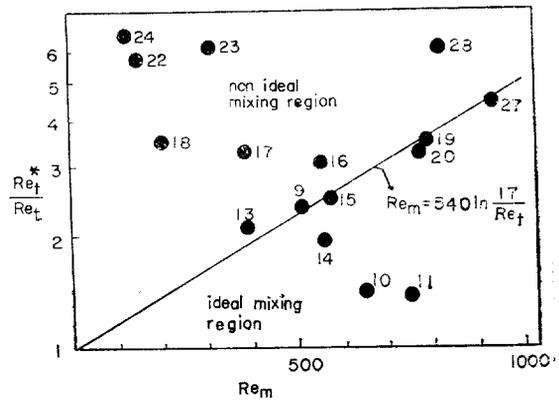


Fig. 9. Experimental regions for ideal and non-ideal mixing.

ical residence time distribution functions, we get the following representative models for the reactor adopted in this study:

Experiment No. 9: Dead region and bypassing model

Fraction bypassing: 0.2
 Fraction of the dead space: 0.26

Experiment No. 24: CSTR and PFR in series model

Volume fraction of CSTR: 0.7

Volume fraction of PFR: 0.3

Experiment No. 45: Dead region without bypassing

Volume fraction of dead space: 0.165

Volume fraction of CSTR: 0.835

Conclusion

1. The minimum value of the tank Reynolds number for perfect mixing for the CFSTR used in the present work was found to be 17.
2. The tank Reynolds number and the impeller Rey-

nolds number for perfect mixing were correlated to be $Re_m = 540 \ln \frac{17}{Re_t}$.

3. For the non-ideal flows, the qualitative description of the mixing patterns is possible with *a priori* mixed models by comparing the results from the tracer experiments with the theoretical RTD of the mixed models.

Nomenclature

- C concentration
- C_F concentration of the feed
- C_0 initial concentration
- D_t tank diameter
- d_m impeller diameter
- $I(\theta)$ internal residence time distribution
- G mass velocity
- m fraction of total volume which is perfectly mixed
- n fraction of feed entering the zone of perfect mixing
- N stirring rate of impeller
- P fraction of feed going to the stagnant zone
- Q flow rate
- Q_a flow rate entering the perfect mixing zone
- V tank volume

- V_a tank volume in perfect mixing zone
- Re_t Reynolds number based on tank diameter
- Re_m Reynolds number based on impeller diameter

Greek Symbols

- ρ density
- μ viscosity
- τ mean residence time
- θ dimensionless time

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