Accepted Manuscript

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PII: S0263-8762(16)30099-5
DOI: <http://dx.doi.org/doi:10.1016/j.cherd.2016.05.016>
Reference: CHERD 2283

To appear in:

Received date: 8-7-2015
Revised date: 29-2-2016
Accepted date: 18-5-2016

Please cite this article as: Ayass, W.W., Nasir, E.F., Farooq, A., Sarathy, S.M., Mixing-Structure Relationship in Jet-Stirred Reactors, *Chemical Engineering Research and Design* (2016), <http://dx.doi.org/10.1016/j.cherd.2016.05.016>

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$$C_{\text{out}}(t)/C_0 = e^{(-t/\tau)}$$

- The effect of geometrical variations in jet-stirred reactors (JSRs) on mixing is studied.
- Monitoring the exit concentration of a tracer using laser absorption spectroscopy collects residence time distribution data.
- Three JSR designs approach the behavior of the ideal reactor model.

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Mixing-Structure Relationship in Jet-Stirred Reactors

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KEYWORDS: Jet-Stirred Reactor, Mixing, Reactor Design, Residence Time Distribution

ABSTRACT: In this study, measurements were performed to assess the overall mixing in jet-stirred reactors (JSRs) passively agitated by feed nozzles. The reactor diameter, nozzle shape, and nozzle diameter were varied to determine the effects of these geometrical parameters on mixing. The mixing was studied at ambient conditions using laser absorption spectroscopy to follow the exit concentration of a tracer gas, carbon dioxide, after a step change in its input flow. The results indicate that the use of a JSR of diameter $D=40$ mm, having inclined or crossed nozzles of diameter $d=1$ mm is recommended for low residence times up to 0.4 sec, while at moderate/high residence times 0.5-5 sec the use of a JSR of $D=56$ mm and $d=0.3$ mm having crossed nozzles is suggested.

Introduction

The ultimate goal of combustion engineering is to reduce pollutant emissions (NO_x , SO_x , etc) and increase efficiency. Consequently, understanding the complex nature of fuel combustion is essential, and working with a fuel that is fully understood is a crucial requirement for engine manufacturers to properly design their systems. This can be achieved either through experimentation or by developing predictive models that are validated against fundamental experiments. In order to validate chemical kinetic models, experiments are performed under ideal conditions – in a jet-stirred reactor (JSR) for example – over a wide range of physical conditions including different ranges of temperature, pressure, and equivalence ratios.

JSRs are commonly used systems to study the oxidation characteristics of gaseous or prevaporized liquid fuels in high and low temperature combustion processes [1,2]. Similar to an ideal continuously stirred-tank reactor (CSTR), the JSR is designed to behave as an ideal reactor or perfectly mixed reactor wherein reactants are well-stirred, leading to a uniform composition throughout the reactor for the entire reactor residence time. Several configurations of JSRs having various injector designs and reactor geometries have been adopted in literature [3-7]. The geometrical rules of designing an ideal JSR are well known [8]. While JSRs are widely applied to validate kinetic models and are almost consistently simulated as kinetically controlled perfectly mixed reactors, there may be situations in which the ideal reactor assumption is invalid. Therefore, due to the lack of residence time distribution (RTD) data in JSRs, our goal was to perform tracer experiments in these reactors to compare with the ideal stirred tank reactor data and to assess the relationship between the nozzle/reactor configuration and the mixing behaviour.

Experimental Procedure

The JSRs used in this study are the most commonly used nowadays in addition to some geometrical variations (Table.1). Configuration **1** ($D=40$ mm, $d=1$ mm, $V=31.6\text{cm}^3$, inclined nozzles) adopted from Dagaut et al. [9] was

manufactured. Configuration **2** is a modified version of **1** but with crossed nozzles, as such nozzle configuration has been adopted from Matras and Villermaux et al. [7]. Configuration **3** ($D=56$ mm, $d=0.3$ mm, $V=90\text{cm}^3$ crossed nozzles) is the original reactor used by Herbinet et al. [10]. Configurations **4**, **5** are manufactured similar to configurations **1** and **2**, respectively, albeit with a smaller nozzle diameter $d=0.3$ mm instead (See Fig.S1-2).

Table 1. A summary of the JSRs design criteria studied. *a:* Calculations are based on the JSR Design rules [8], here the calculations at high temperature are based on: argon as a the flowing gas at 723 K and 10^5 Pa, $A=0.785$, the specific weight and the dynamic viscosity of argon used are equal to $0.71\text{ kg}\cdot\text{m}^{-3}$ and $4.23\cdot 10^{-5}\text{ Pa}\cdot\text{s}$ respectively.

Configuration	D_{Reactor} (mm)	d_{Nozzle} (mm)	Nozzle shape	Residence time range at ambient and high ^a temperature
1	40	1	Inclined	0.03-1.25 sec and
2	40	1	Crossed	0.02-0.46 ^a
3	56	0.3	Crossed	1.2-11.4 sec and 0.5-5sec ^a
4	40	0.3	Inclined	0.33-4.15 sec
5	40	0.3	Crossed	and 0.2-1.53 ^a

The experiments were performed at ambient conditions (298 K, 1 atm), conditions at which mixing studies are usually performed [3,7]. The mixing was studied using a tracer-decay technique (negative step-input mode [11]) using carbon dioxide, CO_2 . The reactor is first filled with CO_2 and is allowed to reach steady-state. A solenoid valve sharply cuts off the CO_2 flow and enables a continuous flow of nitrogen with different constant volumetric flow rates ranging from 18 to 180 cc/sec depending on the desired residence time computed using the volume of the reactor (Table.S1). The decay of CO_2 concentration was monitored at the outlet of the reactor (just at the outlet of the spherical part) by laser absorption spectroscopy (LAS) in the $\nu_1+\nu_3$ band of CO_2 at 2.7 μm . The R26 transition at 3733.47 cm^{-1} is utilized for laser absorption as it is free from interferences due to H_2O transitions [12] (See Fig.S3 and SI for more details about the experimental set-up).

HITRAN simulations were performed in order to obtain expected absorbance levels of CO_2 and interferences from ambient air. Figure 2 shows the frequency range that can be covered by the laser used. This range has lines from the $\nu_1+\nu_3$ band of CO_2 . The R(26) line was used based on expected

interference predicted by the HITRAN simulation. Lines adjacent to R(26) showed some interference from ambient water vapor which would become significant as the CO₂ in the JSR is purged. A fixed wavelength strategy was used for higher temporal resolution and ease in data processing. With the detector bandwidth of 10 MHz and a sampling rate of 2.5 MS/s, the sensor's effective maximum temporal resolution is about 0.4 microseconds. For cases where the residence time was long, the sampling rate was reduced to allow longer recording period by the Data Acquisition System (DAQ card).

Figure 1. A plot of CO₂ and H₂O absorbance versus a frequency range (cm⁻¹). The path length (L) of absorbing species is taken to be as the JSR outlet (~1 cm) and a path length of 7 cm represents the distance where the laser is present in ambient air.

Results and Discussion

If the reactor were to be ideal or perfectly mixed, the decay in tracer concentration would be expressed as follows [11,13]:

$$C_{out}(t)/C_0 = e^{-(t/\tau)} \quad \text{eq.(1)}$$

Where $C_{out}(t)$ is the outlet concentration of the tracer at time t after cutting off the tracer flow, C_0 is the initial steady-state

concentration of the tracer, and τ is the ideal residence time or average residence time [3] – which is equal to the reactor volume over the volumetric flow rate. The normalized residence time distribution (RTD) function $E(t/\tau)$ can be therefore derived from eq. (1) to be expressed as follows [11,13-15],

$$E(t/\tau) = \tau E(t) = e^{-(t/\tau)} \quad \text{eq.(2)}$$

Where, $E(t)$ is the amount of tracer in the reactor over a specified time period which can be derived from eq. (1), and t/τ is the normalized time with respect to the ideal residence time.

As previously mentioned, the experiments were performed at ambient conditions and a fixed wavelength absorption strategy was used. Therefore, the absorbance is directly proportional to the mole fraction and hence to the concentration of the absorbing gas according to Beer's law. Since the relative concentration of the gas is needed for determining the residence time, the relative absorbance from the LAS is sufficient. As expected, the recorded signal increases with time since the CO₂ is being purged. Then, the signal is transformed to a relative absorbance normalized from 1 to 0, that shows an exponential decay with time. The obtained absorbance curves show a nearly ideal exponential decay, so the data was fitted using the ideal model eq. (1) to determine the experimental residence times (See Table.2, Fig.S4-7, SI for more details). Finally, the experimental and ideal RTD curves were calculated by inputting the experimental and ideal residence times in eq. (2) respectively (Fig. 2). Note that the tested residence times were within the maximum and minimum possible operating residence times governed by the JSR design rules [8]. The choice was also dictated by the residence times commonly used in literature in combustion experiments (see Table.S2).

When keeping the volume constant and decreasing the nozzle diameter (configuration 4, 5), the experimental RTD curves of 4 and 5 deviates from ideality. However, it is observed that the crossed configuration in a bigger reactor volume (configuration 3) improves the mixing and its experimental RTD curves coincide with the ideal one at moderate-high residence times. The mixing of the latter deviates from ideality at 0.5 sec because at ambient conditions the operating residence time is 1.2-11.4 sec (Table 1). We should keep in mind that at higher temperatures these operating conditions would change to 0.5-5 sec [10].

Figure 2. $E(t/\tau)$ plots of the different JSRs against normalized time. The upper and lower dashed lines – or error bars – represent the region containing all the experimental data. The error bars of the experimental $E(t/\tau)$ are generated from the error propagation in measuring the residence time in triplicate. The error bars of the ideal $E(t/\tau)$ are sourced from the uncertainty in the volumetric flow rate (i.e., 1% of the mass flow controller's full scale).

The purpose of using a normalized RTD is to enable a valid and direct comparison of fluid flow inside reactors of different sizes and geometries [13]. In other words, when $E(t/\tau)$ is used then *all* perfectly mixed reactors have numerically the same RTD. Interestingly, not all the fitted experimental RTD curves match with the ideal case and it is directly observed that configuration 1, 2 and 3 have one of the best RTDs compared to the ideal one. Looking at configuration 1 and 2, both show an agreement with the ideal mixing behavior suggesting that the mixing in these JSR design specifications is independent from the nozzle shape. Table 2 further confirms this observation by showing a ratio (r) of the experimental residence time to the ideal residence time close to 1. It was reported that the JSR of configuration 1 has a good macromixing at ambient conditions and at 10 atm [7,9] between 0.01-3 sec. According to calculations in Table 1, the range of allowable operating residence time is 0.03-1.25 sec at ambient conditions and 0.02-0.46 sec at 723 K respectively. A computational fluid dynamics (CFD) study [16] suggests that this reactor is suitable for studying the combustion kinetics since the mixing level is ideal or nearly 100%. It is reported that this reactor has a mixing level of around 94 % at a residence time of 0.16 sec, which decreases steadily to reach 93% at a residence time of 0.4 sec [16]. This decrease in mixing is expected since the residence time would be closer to the maximum allowable residence time that is dictated by the condition of having a turbulent flow provided by the jets.

The experimental residence times obtained is observed to always be larger than the ideal case volumetric flow exiting the reactor (Q_{out}) is less than the initial flow entering the reactor (Q_0), which can be computed from the ratio r and confirmed by measuring the actual JSR. The JSR is designed to provide very intense internal recycle streams provided by nozzles, which increases with decreasing the nozzle diameter d or with increasing the JSR radius R [8]. As an example, for configuration 1 at 0.7 sec, r is equal to 1.035, which is very close to ideality; therefore the outflow can be computed to be 96.6% of the initial flow. When keeping the volume constant and decreasing the nozzle diameter d (configuration 4, 5), r is observed to increase diverging from ideality as the RTD plot also shows. This can be correlated to the increased recirculation due to the decrease of d , which increases the residence time of the tracer and thus decreases the outflow by delaying it. The analysis is comparable to the bypassing model of the CSTR where the flow bypasses the reactor thus decreasing the reactor inflow and increasing the actual residence time. In this case, the inflow is less than the outflow due to the intense recirculation streams.

Table 2. The experimental residence time (τ_{exp}) compared to the ideal residence time (τ_{id}), their ratio ($r = \tau_{exp}/\tau_{id}$) and the

percent ratio of the volumetric flow rate exiting the reactor ($100\% \cdot Q_{out}/Q_0$) of configuration 1-5.

Configuration	τ_{id}	$\tau_{exp.}$	τ_{exp}/τ_{id}	Q_{out}/Q_0
1	0.37	0.415 $\pm$ 0.011	1.120	89.30
	0.7	0.725 $\pm$ 0.027	1.035	96.60
2	0.37	0.402 $\pm$ 0.073	1.080	92.60
	0.7	0.722 $\pm$ 0.013	1.031	97.00
3	0.5	0.678 $\pm$ 0.025	1.356	73.70
	2.5	2.630 $\pm$ 0.100	1.052	95.00
	5	5.530 $\pm$ 0.180	1.106	90.00
4	0.7	0.902 $\pm$ 0.070	1.289	77.6
	1	1.160 $\pm$ 0.053	1.160	86.2
5	0.37	0.493 $\pm$ 0.012	1.330	75.20
	0.7	0.940 $\pm$ 0.082	1.340	75.00
	1	1.700 $\pm$ 0.0812	1.70	58.80

Conclusions

The present experimental results and calculations on mixing in JSRs, confirm what is found in the literature, and adds important observations regarding the JSR design-mixing relationship. We have demonstrated, for the first time, the use of LAS to determine the operating residence time range for various JSR configurations ensuring an overall well-mixed environment. We therefore recommend using the JSR of configuration **1** or **2** only at short residence times of 0.02-0.4 sec under combustion conditions, although this reactor still performs ideally at 0.7 sec at ambient conditions. However, when a larger residence time ranging between 0.5-5 sec is required, then configuration **3** is recommended. While of importance, mixing is only one side ensuring good experiments and the second important criterion is thermal homogeneity. Both mixing and thermal homogeneity in such reactors should be evaluated at elevated temperatures and pressures to determine how close they behave to ideality.

ASSOCIATED CONTENT

Supporting Information Available: JSR images and designs, Experimental set-up details and scheme, Sample raw data and calculations, Experimental residence times, Commonly used residence times in combustion experiments.

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ACKNOWLEDGMENT

This work was supported by the King Abdullah University of Science and Technology Clean Combustion Research Center and Saudi Aramco under the FUELCOM program. We thank Dr. *Frédérique* Battin-Leclerc, Dr. Olivier Herbinet, and Dr. Philippe Dagaut for providing jet-stirred reactor designs. We also thank Dr. Ghada El-Kadamany and Rachelle Smith for their help in proofreading the manuscript.

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