

## THE ENHANCEMENT OF ULTRASOUND REACTOR DESIGN USING TRACER METHODOLOGY

Mircea-Teodor NECHITA, Elena Niculina DRĂGOI, Adrian Cătălin PUIȚEL, Gabriel Dan SUDITU\*

*Technical University “Gheorghe Asachi” of Iași, Faculty of Chemical Engineering and Environmental Protection “Cristofor Simionescu”, Bd. Prof. Dimitrie Mangeron, No. 73, 700050, Iași, România, \*gabriel-dan.suditu@academic.tuiasi.ro*

**Abstract:** *In this work a hydrodynamic study over an ultrasound reactor was performed using the tracer methodology. The occurrence of flow short-circuits and dead-zones was demonstrated using two of the residence time distribution functions  $C(t)$  and  $E(t)$ . In order to improve the reactor design two baffles were added to its construction. The presence of the baffles significantly affects the fluid flow, increasing the residence time and the degree of mixing inside the reactor.*

**Keywords:** *ultrasound reactor, residence time distribution, fluid flow, short-circuits, baffles*

### 1. Introduction

The concept of “sonochemistry” generally describes the overall chemical and physical processes that take place in a solution exposed to a sonic field in the range from 20 kHz to 2 MHz [Colmenares,2017]. Even though a large number of applications of sonochemistry are reported in the literature [Colmenares,2017; Mason,2001; Pokhrel,2016; Rosca,2003], the design and optimization of ultrasound reactors also known as sonochemical reactors or as cavitation reactors are still open to progress [Cravotto,2005; Gogate,2003; Gogate,2004; Dong,2020; Asgharzadehahmadi,2016; Sutkar,2009]. The transducer type and number, the transducer cooling system, the ratio of vessel/transducer diameters, the frequency of the sound field, the liquid height, the flowrate and the liquid temperature are among the parameters that must be considered when designing ultrasonic reactors [Sutkar,2009; Wood,2017; Asakura,2008; Gogate,2000]. Besides these specific parameters, the reactor performances are highly influenced by the efficiency of the mass transfer, which is directly depending on the mixing time and flow pattern [Asgharzadehahmadi,2016; Dong,2017].

It is a common practice to use commercial ultrasound (US) apparatus (baths, cleaners, sterilizers) as ultrasonic emitters for sonochemical reactors [Cravotto,2005; Romdhane,1997] but this comes with a series of design constraints related with the shape, size and position of input/output pipes that significantly affects the fluid flow through the reactor and consequently the reactor performances. Therefore, the understanding the flow patterns inside any reactor is essential for the engineering design. It is even more important for photochemical, sonochemical or microwave reactors where the homogeneity of the radiation exposure is required. From the engineering point of view, the residence time distribution methodology (RTD) is one of the most enlightening characterizations of the fluid flow in a reactor [Sans,2012]. It is fast, cheap, simple and easy to elucidate and supplies valuable information about so called flow “defects”: channeling (short-circuits, by-passing), dead-zones (stagnancies), back-mixing and allows the comparison with some ideal systems [Bérard,2020; Gondrexon,1998].

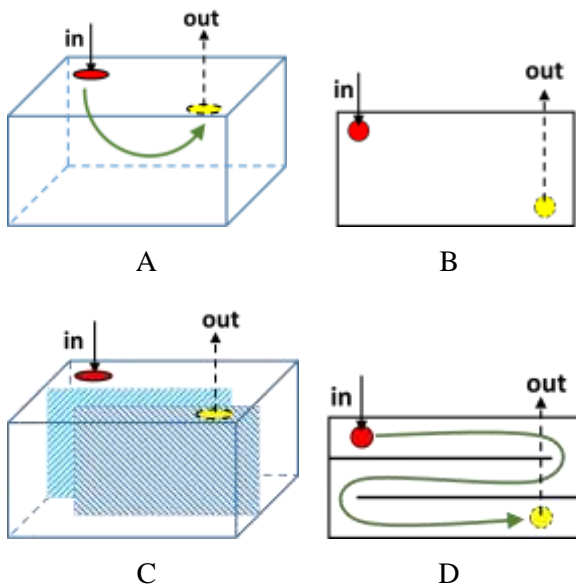
The ultrasound reactor (US-R) under study is a part of a lab-scale prototype that contains a series of three reactors: chemical, ultrasound,

ultraviolet (pending patent), a tampon vessel and a recirculation pump [Suditu,2021]. The tracer methodology (RTD analysis) was used to investigate the fluid flow through the US-R. The first results indicated the presence of short-circuits and dead-zones. In order to improve the reactor design and to increase the residence time, two baffles were added to its constructions. The presence of the baffles significantly affects the fluid flow and the degree of mixing inside the reactor.

## 2. Experimental

### 2.1. Reactor design, input/output location, baffles placement

The ultrasound reactor with rectangular parallelepiped shape and a total volume of 750 cm<sup>3</sup> was made from plexiglass. The input/output pipes were placed on the top of the reactor in opposite corners, as presented in Figs. 1, A, B. In order to increase the fluid path through the reactor two paralleled baffles were placed inside the US-R as presented in Fig. 1, C, D.



**Figure 1:** The ultrasound reactor: side view and top view without baffles (1, 2); side view and top view with baffles (3, 4); the green line: – hypothetic fluid pathways through the reactor, the red circle: input pipe; the yellow circle: output pipe.

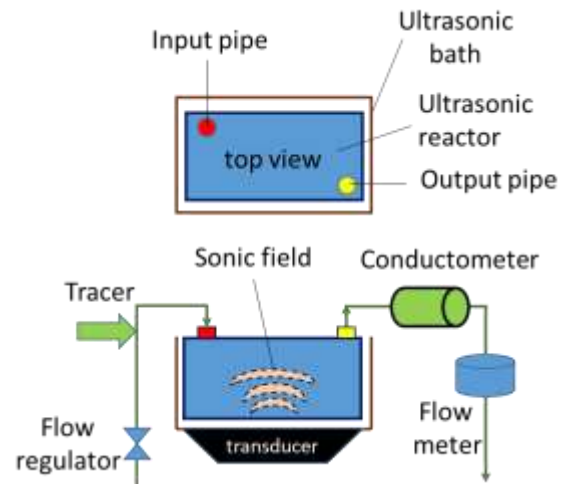
### 2.2. Tracer methodology

A sodium chloride solution of 20% mass

concentration was used as a tracer, the response being registered with a WTW conductivity meter Cond 315i. The conductivity - concentration conversion was made using a calibration curve ( $R^2 = 0.99965$ ) [Nechita, 2019]. The raw experimental data given by the concentration vs time plot (C - curve) were supplementary explored using the residence time distribution function  $E(t)$ . The analysis of the E - curve provide valuable information about the presence of flow defects (“channeling” or “short-circuits” and “stagnancies” or “dead zones”).

### 2.3 Experimental set-up

The experimental set-up is presented in Fig. 2. The US-R was placed into an ultrasonic bath type Ultrasonic Cleaner PS-10A. The water flow rate was controlled using a flow regulator and measured at the exit of the system using a flowmeter. The NaCl solution was inserted with a medical syringe into the silicon pipe, close to the reactor input. The conductometer was placed on the exit pipe, near the reactor output.



**Figure 2.** The experimental set-up for RTD analysis

## 3. Results and discussions

### 3.1 The C - curve

The C - curve gives the time evolution of the tracer concentration and provides the raw data for RTD investigation. Just by observing the C - curves alone, without any supplementary data analysis, the baffle influence is evident. The peak visible at the beginning of the experiments

(Figs. 3 A, B) is a clear evidence of short-circuiting or by-passing, when a considerable part of the tracer passes directly through the US-R, probably without or limited mixing. The long tail that appear in Figs. 3 (A, B) is

commonly related with the presence of stagnancies and/or dead-zones, when a part of the tracer stays in the reactor a prolonged time.

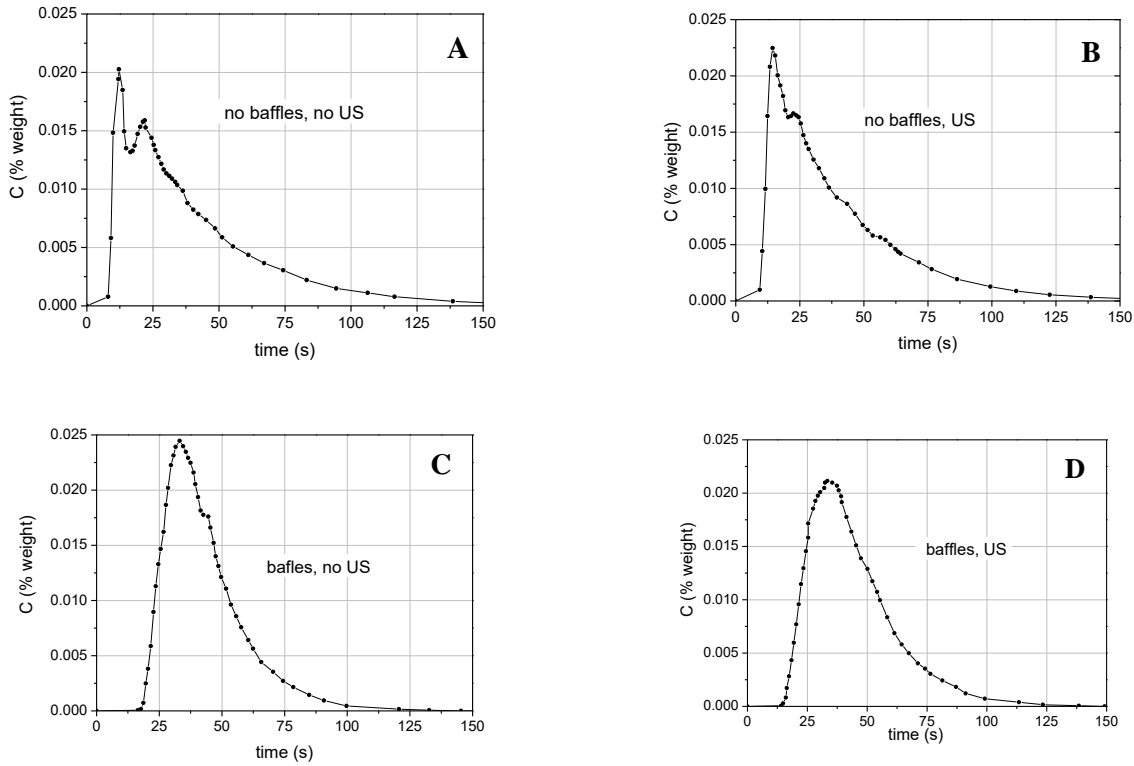


Figure 3 The C curves: A (without baffles, without US); B (without baffles, with US); C (with baffles, without US); D (with baffles, with US)

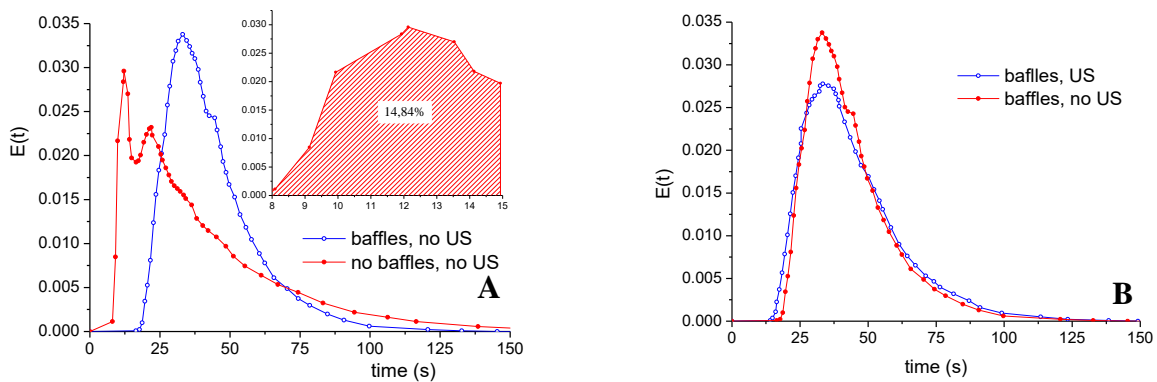


Figure 4. The E – curves: A – without US, without baffles vs. without US, with baffles; B – with baffles, with US vs. without baffles, without US

Due to the reactor geometry and the placement of input/output pipes the most probable position

of the stagnancies are at the reactor's bottom corners.

### 3.2 The E – curve

E(t) - the residence time distribution also known as the exit age distribution is one of the main RTD functions [Sans,2012; Fogler,2016]. It can be appraised using Eq. (1):

$$E(t) = \frac{C(t)}{\int_0^{\infty} C(t) \cdot dt} \quad (1)$$

and its main property is that:

$$\int_0^{\infty} E(t) \cdot dt = 1 \quad (2)$$

The analysis of E – curve allows a relative quantification of the time spent in reactor by the elements of fluid [Fogler,2016]. The examination of the E curve for the reactor without baffles and without US exposure (Fig. 4 A, inset) shows that 14.84% of the material has spent 15 or less than 15 seconds in the reactor. Note that during this time no signal were yet recorded in the case of the reactor with baffles. On the other hand, after 60 seconds, the percent of the tracer passed through was 78.98% for the reactor without baffles and 85.94% for the reactor with baffles. The baffles insertion narrows the residence times for the elements of the fluid. The sonic field also has an influence towards the residence time (Fig. 4 B): 83.81% of the tracer spent 60 or less than 60 seconds inside the reactor with baffles in presence of US, a little lower than 85.94% when the US where not used. The fact that the presence of US faintly widens the residence time (Fig. 4, B) might be related with the mixing effect caused by the sonic vibrations [Wood,2017; Xu,2013 Vichare,2001].

Similar profiles of the E(t) curves, highlighting the presence of short-circuits, were recently reported for a cylindrical sonochemical reactor [Ilewicz,2020].

In order to estimate the degree of mixing (age distribution of the fluid elements) some other RTD functions must be plotted and also the flowrate influence have to be considered [Ilewicz,2020; Toson,2019], and these will be the subjects of our future studies.

### 4. Conclusions

In order to study the flow behavior in a rectangular parallelepiped shape ultrasound reactor tracer investigations were conducted. Two representative RTD functions were plotted as function of time and the results were interpreted following the RTD methodology. Due to the constructive restrains of the reactor the presence of flow defects such as short-circuits and dead zones cannot be avoided. In order to enhance its design two baffles were placed inside the reactor. The RTD analysis shows that the presence of the baffles significantly affects the fluid flow, increase the residence time and the degree of mixing inside the reactor.

### Acknowledgements

This work was supported by project PN-III-P4-ID-PCE no 58/2021 financed by UEFISCDI, Romania

### 4 References

- [Colmenares,2017] Colmenares, J.C., Chatel, G., *Sonochemistry: from basic principles to innovative applications*, Springer, 2017.
- [Mason,2001] Mason, T., Tiehm, A., *Advances in sonochemistry, ultrasound in environmental protection*, (Vol. 6), 2001.
- [Pokhrel,2016] Pokhrel, N., Vabbina, P.K., Pala, N., *Sonochemistry: Science and Engineering. Ultrasonics Sonochemistry*, 29, 104-128, 2016.
- [Rosca,2003] Rosca, I., Nechita, M.T., Apostolescu, G.A., *Sonochemistry and the chemical effects of the ultrasound, Buletinul Institutului Politehnic din Iași, Chimie și Inginerie Chimică*, XLIX (LIII), (5), 51-56, 2003.
- [Cravotto,2005] Cravotto, G., Omiccioli, G., Stevanato, L., *An improved sonochemical reactor. Ultrasonics Sonochemistry*, 12, (3), 213-217, 2005.
- [Gogate,2003] Gogate, P.R., Wilhelm, A.M., Pandit, A.B., *Some aspects of the design of sonochemical reactors, Ultrasonics Sonochemistry*, 10, (6), 325-330, 2003.

7. [Gogate,2004] Gogate, P.R., Pandit, A.B., Sonochemical reactors: scale up aspects, *Ultrasonics Sonochemistry*, 11, (3), 105-117, 2004.
8. [Dong,2020] Dong, Z., Delacour, C., Mc Carogher, K., Udepurkar, A.P., Kuhn, S., Continuous Ultrasonic Reactors: Design, Mechanism and Application, *Materials*, 13, (2), 344, 2020.
9. [Asgharzadehahmadi,2016] Asgharzadehahmadi, S., Abdul Raman, A.A., Parthasarathy, R., Sajjadi, B., Sonochemical reactors: Review on features, advantages and limitations, *Renewable and Sustainable Energy Reviews*, 63, 302-314, 2016.
10. [Sutkar,2009] Sutkar, V.S., Gogate, P.R., Design aspects of sonochemical reactors: Techniques for understanding cavitation activity distribution and effect of operating parameters, *Chemical Engineering Journal*, 155, (1), 26-36, 2009.
11. [Wood,2017] Wood, R.J., Lee, J., Bussemaker, M.J., A parametric review of sonochemistry: Control and augmentation of sonochemical activity in aqueous solutions, *Ultrasonics Sonochemistry*, 38, 351-370, 2017.
12. [Asakura,2008] Asakura, Y., Nishida, T., Matsuoka, T., Koda, S., Effects of ultrasonic frequency and liquid height on sonochemical efficiency of large-scale sonochemical reactors, *Ultrasonics Sonochemistry*, 15, (3), 244-250, 2008.
13. [Gogate,2000] Gogate, P.R., Pandit, A.B., Engineering design method for cavitation reactors: I. Sonochemical reactors, *AIChE Journal*, 46, (2), 372-379, 2000.
14. [Dong,2017] Dong, Z., Zhao, S., Zhang, Y., Yao, C., Yuan, Q., Chen, G., Mixing and residence time distribution in ultrasonic microreactors, *AIChE Journal*, 63, (4), 1404-1418, 2017.
15. [Romdhane,1997] Romdhane, M., Gadri, A., Contamine, F., Gourdon, C., Casamatta, G., Experimental study of the ultrasound attenuation in chemical reactors, *Ultrasonics Sonochemistry*, 4, (3), 235-243, 1997.
16. [Sans,2012] Sans, V., Karbass, N., Burguete, M.I., García-Verdugo, E., Luis, S.V., Residence time distribution, a simple tool to understand the behaviour of polymeric mini-flow reactors, *RSC advances*, 2, (23), 8721-8728, 2012.
17. [Bérard,2020] Bérard, A., Blais, B., Patience, G.S., Experimental methods in chemical engineering: Residence time distribution-RTD, *The Canadian Journal of Chemical Engineering*, 98, (4), 848-867, 2020.
18. [Gondrexon,1998] Gondrexon, N., Renaudin, V., Petrier, C., Clement, M., Boldo, P., Gonthier, Y., Bernis, A., Experimental study of the hydrodynamic behaviour of a high frequency ultrasonic reactor. *Ultrasonics Sonochemistry*, 5, (1), 1-6, 1998.
19. [Suditu,2021] Suditu G.D., Nechita M.T., Puițel A.C., Drăgoi E.N., *Instalație pentru epurarea apelor uzate prin metode foto-sonochimice*, Patent number RO135064-A2, 2021.
20. [Nechita,2019] Nechita, M.T., Drăgoi, E.N., Suditu, G.D., Puițel, A.C., in Analysis of the Residence Time Distribution of a Photochemical Reactor, *21<sup>st</sup> Romanian International Conference on Chemistry and Chemical Engineering*, RICCCCE 21, Constanța, România, 4-7, 2019.
21. [Fogler,2016] Fogler, H.S., *Elements of chemical reaction engineering*, Prentice Hall, 2016.
22. [Xu,2013] Xu, Z., Yasuda, K., Koda, S., Numerical simulation of liquid velocity distribution in a sonochemical reactor, *Ultrasonics Sonochemistry*, 20, (1), 452-459, 2013.
23. [Vichare,2001] Vichare, N.P., Gogate, P.R., Dindore, V.Y., Pandit, A.B., Mixing time analysis of a sonochemical reactor, *Ultrasonics Sonochemistry*, 8, (1), 23-33, 2001.
24. [Ilewicz,2020] Ilewicz, W., Skupin, P., Choiński, D., Błotnicki, W., Bielecki, Z., On-Line Estimation of the Ultrasonic Power in a Continuous Flow Sonochemical Reactor, *Energies*, 13, (11), 2952, 2020.
25. [Toson,2019] Toson, P., Doshi, P., Jajcevic, D., Explicit Residence Time Distribution of a Generalised Cascade of Continuous Stirred Tank Reactors for a Description of Short Recirculation Time (Bypassing). *Processes*, 7, (9), 615, 2019.