



Static Mixers for Water Ozonation: Applications and Mathematical Modelling - A Review

*Mohamed A. M. Saad, Abdullah. A. Elamari, Awad. E. Alshebani, E. M. Elmabrouk, Sana Ibrahim Abukanisha

Department of Chemical Engineering, Faculty of Engineering, University of Sirte, Libya

*Corresponding author: mohammed.saad@su.edu.ly

Abstract Static mixers have been successfully employed in water and wastewater treatment, particularly in water ozonation for disinfection and oxidation purposes. Producing higher concentration of ozone requires new contactors that operate efficiently at low gas/liquid ratio. The Kenics static mixer can meet these requirements and therefore enhance the ozone mass transfer rate. This paper summarises the field of static mixers in water ozonation including comparison with other gas/liquid contactors. This study also reviews recent conceptual and technological innovations in static mixers: Current industrial applications, advantages and types are discussed, focusing on mixing and mass transfer performance. The work is complemented by a review of mixing fundamentals, knowledge of which allows the development of mathematical models which are crucial for the analysis of experimental data. Moreover, it reviews the recent advances of the mathematical models of zone mass transfer in static mixer: back flow cell model (BFCM), Axial diffusion model (ADM) and the continuous flow stirred tanks in series (CFSTR's in series). Both the steady state and the transient BFCMs were validated with experimental data by Tizaoui and Zhang and it was found that they accurately predicted the outlet ozone concentration and the impulse RTD curves of gas-liquid system respectively along height of the static mixer.

Keywords: Gas liquid contactors, Mathematical Modelling, Static Mixers.

استخدام تقنية الخلطات الثابتة في تعقيم المياه بالأوزون: التطبيقات، النماذج الرياضية-نبذة عن الدراسات السابقة

*محمد عبدالسلام محمد سعد و عبدالله على العماري و عوض امحمد الشيباني و المبروك محمد المبروك ابولبيدة و

سنا ابراهيم ابو كنيشة

قسم الهندسة الكيميائية-كلية الهندسة- جامعة سرت، ليبيا

*المراسلة: mohammed.saad@su.edu.ly

المخلص تم استخدام الخلطات الثابتة بنجاح في معالجة المياه والمياه العادمة، وخاصة تقنية تعقيم المياه بالأوزون وعمليات الأكسدة. يتطلب إنتاج تراكيز عالية من الأوزون استخدام تقنيات تلامس جديدة تعمل بكفاءة عند نسبة منخفضة من الغاز/ السائل. يمكن للخلط الكينيكي الثابت تلبية أو تحقيق هذه المتطلب وبالتالي تعزيز معدل انتقال كتلة الأوزون. تلخص هذه الورقة نبذة عامة عن الدراسات السابقة في مجال عمل الخلطات الثابتة وأهميتها في معالجة المياه بالأوزون متضمنة على مقارنة أجهزة تلامس أخرى نوع غاز/ سائل. كما تستعرض أيضا الابتكارات والتكنولوجية الحديثة في صناعة الخلطات الثابتة مثل التطبيقات الصناعية لأنواع الحالية، كذلك مزاياها والتركيز على عملية الخلط ومعدلات انتقال الكتلة. كما يستكمل هذا العمل بمراجعة أساسيات عملية الخلط، والتي تسمح بتطوير نماذج رياضية تعتبر أساسية في تحليل البيانات التجريبية، علاوة على ذلك، فإن هذه المراجعة تستعرض التطورات الحديثة للنماذج الرياضية لانتقال كتلة الأوزون في خلط ثابت كنموذج (BFCM) - (ADM) و (CFSTR's in series). كذلك تم التحقق من صحة كل من الحالة المستقرة والانتقالية BFCMs من خلال بيانات تجريبية ل (Tizaoui و Zhang) ووجد أنهم توقعوا بدقة تركيز الأوزون الخارج ومنحنيات RTD الدافعة لنظام الغاز السائل على طول ارتفاع الخلط الاستاتيكي.

الكلمات المفتاحية: موصلات التلامس سائل-غاز، النمذجة الرياضية، الخلطات الساكنة.

1. Introduction

A lot of processes in the field of chemical engineering are based on the chemical reaction between the solute gas and soluble species present in liquid. In nature, gas diffuses into the liquid through gas-liquid interface due to the concentration difference of the gas between the liquid at the surface and the liquid bulk without mechanical energy. Because both the gas mass transfer rate and the reaction rate are low, new gas-liquid contactors such as static mixers are used in

order to increase the gas mass transfer rate and therefore faster reaction rate was obtained. These contactors enhance the mass transfer rate by increasing surface area between the gas and liquid and increasing the intensity of turbulence [3,4].

Ozonation is currently used as an alternative disinfection method to the chlorination process in water treatment, it has many drawbacks: it is incompatible with membrane processes and it result in formation of a dangerous

organochlorinated compounds [5]. In water and wastewater treatment, ozone is used as biocide, oxidant and as pre-treatment reagent so as to improve the efficiency of the settling and filtration processes. Ozone is currently widely used because of its ability to kill bacteria, viruses, spores and protozoa. The new stringent standards for drinking water require optimization of the ozonation process through improving the ozone gas mass transfer to the liquid phase [5-7]. The ozonation process is practised by dissolving gaseous ozone into the liquid water so as to react with target contaminants. Water ozonation usually consists of four steps: convection and back mixing of the liquid flowing through contacting chambers inside the static mixer. These two processes occur simultaneously with two other processes: ozone gas mass transfer from gas phase to the liquid phase, ozone decomposition and reactions of ozone with organic material in the water [8].

The ozonation process is an effective method for removing the organic contaminants from water. It does not only depend on the mass transfer of ozone from the gas phase to liquid water, but also on the kinetic reactions of the ozone with the pollutants. The ozone mass transfer efficiency depends on the hydrodynamics of the fluids. However, the decay rate of the ozone depends on pH and temperature [9]. During the last few decades, new cheap ozone generators have been developed and they produce high ozone concentration, but they require low gas flow rates which result in low gas to liquid volumetric flow rate ratio. As a result, classical bubble columns are no longer applicable because they require a larger gas flow rate to achieve efficient mixing between gas and liquid and therefore high ozone mass transfer. Static mixers, however, are more efficient at low gas/liquid ratio and high concentration of ozone and therefore they provide a solution to this problem. Moreover, static mixers produce a homogeneous ozone concentration because of the high turbulence produced by the elements that are inside the mixer, which ensure adequate gas/liquid mixing [1, 2]. Packed towers are extensively used for gas-liquid processes in many industries because of their effectiveness and cost. However, in the last few decades, static mixers have received a great attention because of their characteristics: high void fraction leading to low pressure drop with a very efficient gas-liquid contact and distribution [4]. Since it is difficult to ensure adequate contact between the gas ozone and the liquid water in conventional bubble columns, static mixers have been developed as an attractive alternative in order to enhance the ozone mass transfer rate.

A static mixer, as shown in figure 1, is designed as a series of identical, motionless inserts which are called elements. Its function is to redistribute fluid in a direction transverse to the main flow and in radial and tangential directions [4, 1].



Fig. 1: Kenics static mixer [10]

Static mixers have many advantages over other gas-liquid contactors which can be summarised as follows:

- Small bubble diameter and therefore very high interfacial area.
- High mass transfer coefficient.
- Plug flow.
- Little maintenance and low power consumption since they have no moving parts except the pump.
- Higher performance at low energy consumption.
- Narrow residence time distribution.
- Low equipment cost.
- Most of the factors mentioned above mean higher mixing between the ozone gas and the water enhanced heat and mass transfer [11-13, 34].

Heyouni et al. [14] stated that the performance of a static mixer is better than other conventional gas-liquid contactors such as bubble columns and stirred vessels. This has been represented by Figure 2.

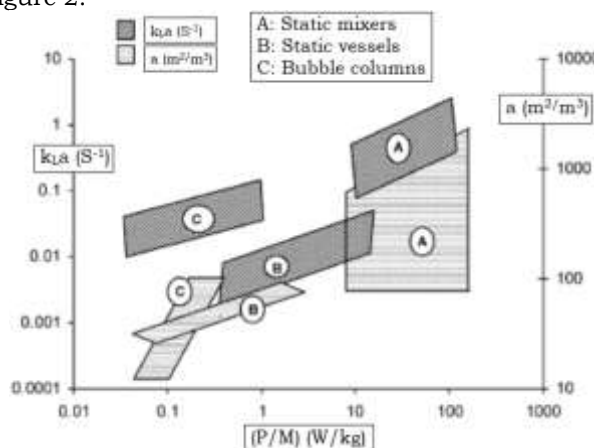


Fig. 2: Comparison of mass transfer coefficient and interfacial area between static mixers and traditional gas-liquid contactors [14]

Almost all the mathematical models that are developed to predict the performance of the ozone contactors are based on one of the following two assumptions: complete mixed flow or plug flow exist in the liquid phase. Applying these assumptions in modelling the gas-liquid contactors will underestimate the performance of the ozone contactor. Because of phenomena of the axial dispersion of the liquid phase, the real flow regime is closer to mixed flow than the plug flow, but it is not perfectly mixed flow. Thus, the back flow cell model (BFCM) has been developed as an alternative way to describe the hydrodynamics and mass transfer of the ozone inside the Kenics static mixer [15]. The BFCM is a general form of stagewise backmixing models and it can be used to characterise the backmixing in the liquid phase for co-current or counter-current gas-liquid contactors at steady state or unsteady state operating conditions [16].

The BFCM is a mathematical model that is applied to characterise the performance of the Kenics ozone static mixer. In order to describe the axial dispersion in the liquid phase, the BFCM assumes a back flow between the cells in the opposite

direction to the main liquid flow and exchange flow in the same direction of the main liquid flow. These two flows have been expressed as back flow ratio (B) and exchange flow ratio (B) and both of them are assumed to be equal and constant along the mixer. Generally, BFCM is composed of two series of equal number of completely mixed cells in which one series describes the liquid phase and the other describes the gas phase. It uses the cells number and back flow ratio to describe the backmixing in the liquid phase [17].

The steady state BFCM has been developed by performing a basic material balance with respect to the ozone was performed around each cell inside the static mixer. The ozone mass transfer rate of ozone to the water and also its reaction in the main bulk of the liquid phase were included in the basic mass balance equations [34].

1.1. Gas-Liquid Reactors

In the case of a gas-liquid chemical reaction, the gas solute (ozone) is absorbed from the gas phase to the bulk of the liquid phase in two sequential steps. Firstly, the gas solute is dissolved into the gas-liquid interface solution. Secondly, the diffusion of the solute through the boundary layer of the solution accompanied with chemical reaction, which control the overall rate of absorption. The reaction might happen between the solute and either the pollutants dissolved in the solution or with the solvent. The absorption rate will be much higher with the presence of chemical reaction than the physical absorption only [18, 19]. Several forms of gas-liquid contactors are listed in the Figure 3 below.

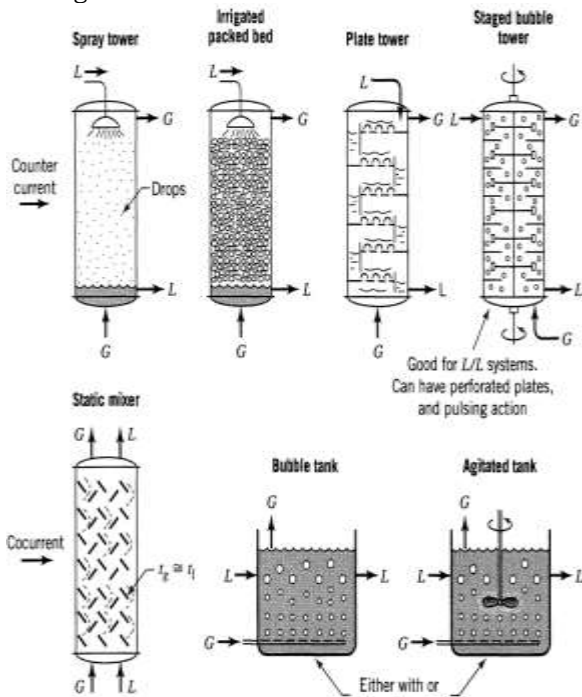


Fig. 3: Different types of gas-liquid contactors [18]

The mass transfer in the gas-liquid reactor occurs during either the physical absorption of the gases into the liquid phase or the chemical reaction between the absorbed gas and another solute present in the liquid phase.

1.1.1. Physical absorption

It is the process in which the solute is transferred from the gas phase to the liquid phase without reacting with species present in the liquid phase. Lewis and Whitman, [19] have postulated the double film theory to describe the physical absorption process.

1.1.2. Two film theory of mass transfer

Whitman [19] postulated that when a gas and liquid are in contact, the gas and liquid main bulk are in turbulent flow and uniformly mixed because of mass transfer by convection. Moreover, he suggested that there are two stagnant films of gas and liquid adjacent to the interface through which the absorbed gas is transported by molecular diffusion as shown in Figure 4.

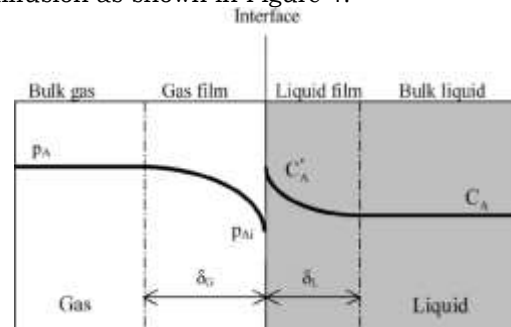


Fig. 4: The double film theory [19]

The gaseous solute A is transported from the gas bulk through the interface and then to the liquid bulk. The species (A) has given solubility, which depends on the difference in the solute concentration between the two phases.

For the gas film:

$$N_A = k_{Ag}(P_A - P_{Ai}) \tag{1}$$

Where:

k_{Ag} is gas film mass transfer coefficient.

P_A and P_{Ai} are the dissolved gas pressure in the liquid bulk and at the interface respectively [19].

By applying the same procedure to the transport of A through the liquid film:

For the liquid film:

$$N_A = \frac{D_{Al}(C_{Ai} - C_A)}{\delta_l} = k_{Al}(C_{Ai} - C_A) \tag{2}$$

Where:

k_{Al} is liquid film mass transfer coefficient.

C_A and C_{Ai} are the dissolved gas concentration in the liquid bulk and at the interface respectively [19].

1.1.3. Henry's law

The gas and liquid at the interface are assumed to be in a state of equilibrium. This means that there is not resistance to mass transfer at the interface. The equilibrium relationship has been described by Henry's law:

$$P_{Ai} = H_A C_{Ai} \tag{3}$$

Where:

H_A : is the Henry's law constant of the solute A [18, - 20].

There are three special cases depending on the value of the of k_{Ag} and k_{Al} [20]:

Gas solute is highly soluble

When the solute is very soluble gas, the Henry's law constant (H_A) is very large. Therefore, the system is gas film controlled. This can be demonstrated by the following equations:

$$K_{AG}(P_A - P_A^*) \approx k_{Ag}(P_A - P_{Ai}) \quad (4)$$

The above equation can be written as:

$$K_{AG} \approx k_{Ag} \quad (5)$$

Gas solute is slightly soluble

When the ozone gas is sparingly soluble, the value of H_A is very small. Thus, the resistance to mass transfer is confined in the liquid phase (liquid film controlled), which can be represented by the following expression:

$$K_{AL} \approx k_{Al} \quad (6)$$

The resistance met by the soluble gas A is due to the collision between the molecules of the solute and the interfering molecules of the gas or the liquid bulk through which the diffusion occurs.

Gas solute is intermediately soluble

The resistance of both films are important and they should be considered in the design of mass transfer equipment.

1.1.4. Mass transfer with chemical reaction

The main objective of gas-liquid mixing processes accompanied with chemical reaction is to provide a gaseous reagent which reacts with other contaminants in the solution or with solvent as it dissolves or after being absorbed into the solution. In such processes, the chemical reactions are mainly divided into: slow chemical reactions that occur in the liquid bulk after gas absorption. Secondly, fast chemical reaction which is mostly or completely takes place in the liquid film close to the gas-liquid interface as the gaseous solute dissolves [19, 20].

The gas absorption in the liquid phase accompanied with chemical reaction is faster than that of pure physical absorption at the same driving force. This has been attributed to the influence of the chemical reaction on the concentration of the gas in the vicinity of the gas-liquid interface. It is acknowledged that slow reactions reduce the concentration of the gaseous solute in liquid bulk, whereas fast reactions consume all the gaseous solute in the liquid bulk resulting in higher driving force for mass transfer. Moreover, increasing the reaction rate will lead to a higher mass transfer coefficient. This has been attributed to the fact that the chemical reaction produces an effective reaction layer of thickness δ_R which is smaller than the thickness of the diffusion layer δ_L for the physical absorption. Therefore, Hatta number and enhancement reaction factor has been developed to represent the effect of the chemical reaction [3, 19, 20].

1.1.5. Enhancement factor

Fast chemical reactions have strong influence on the liquid side mass transfer coefficient. However, the gas phase mass transfer coefficient remains the same because the chemical reaction occurs only in the liquid phase. This reaction enhances both the liquid mass transfer coefficient and the driving force for absorption. It should be noted that the absorption rate of the solute accompanied with chemical reaction is larger than that of pure physical absorption. Therefore, the enhancement

factor, E has been used to account for the effect of the chemical reaction [20, 21].

$$N_A = Ek_1C_{Ai} \quad (7)$$

This can be compared to equation mentioned earlier for the pure physical absorption:

$$N_A = k_{Al}(C_{Ai} - C_A) \quad (8)$$

The enhancement factor can also be presented as the ratio of the rate of transfer with chemical reaction to the rate of transfer without chemical reaction:

$$E = k_1/k_l \quad (9)$$

1.1.6. Hatta number (Ha)

Hatta number is a measure of the quantity of the gas solute that reacts in the diffusion layer adjacent to the gas-liquid interface in comparison with the amount of solute which dissolves into the liquid bulk without reacting and it has the following formula:

$$\begin{aligned} Ha^2 &= \frac{\text{maximum possible conversion in the film}}{\text{maximum diffusion transport through the film}} \\ &= \frac{k_C D_{Al}}{k_{Al}^2} \end{aligned} \quad (10)$$

Where:

Ha^2 is Hatta number

k_C is the modified reaction rate constant.

D_{Al} is the diffusion time, and known as Damkoher number.

k_{Al}^2 is the individual mass transfer coefficient of the solute in the liquid bulk.

The role of the Hatta number is tell whether the reaction is slow, intermediate or fast. When the Hatta number is much greater than one, this means all reaction takes place in the film and gas-liquid interfacial area is the controlling factor [20, -22].

However, if $Ha \ll 1$, no reaction occurs in the film and the bulk volume controls the rate. Thus, depending on the magnitude of the Hatta number, we will determine whether the reaction occurs completely or partly in the liquid bulk as shown below:

1. If $Ha < 0.02$, the reaction is infinitely slow.
2. If $0.02 < Ha < 2$, we have intermediate reaction.
3. If $Ha > 2$, the reaction is fast and happens in the film [18].

1.1.7. Slow chemical reaction in the main body with film resistance

This is very important case, where all the reaction takes place in the main body of the liquid. However, the film resistance to mass transfer still exists. This process begins with an almost pure physical absorption and end up with a chemical reaction in the main body of the liquid as shown in Figure 5. If the reaction is fast enough, the concentration of A (C_A) in the liquid bulk is negligible and the enhancement factor is set to 1 [18, 19].

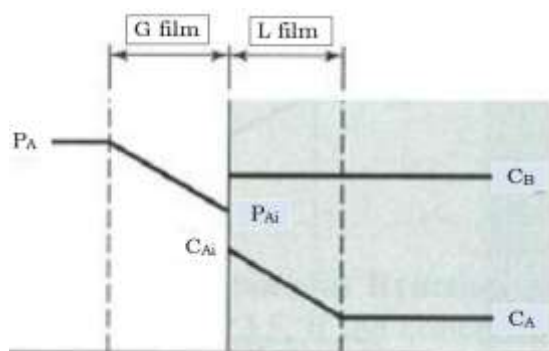


Fig. 5: Shows slow reaction with the film resistance [18]

The reaction rate is utterly determined by the mass transfer through the liquid film and increases with increasing the liquid mass transfer coefficient ($k_L a$)

$$R = k_{La} C_A \quad (11)$$

Large interfacial area and high liquid hold up are needed for this process. These two requirements can be met by using static mixers [11].

2. Static Mixers

Static mixers have been established in the process industry since 1970s. There are about 2000 US patent and more than 8000 articles about the static mixers and their uses. Around 30 commercial models are presently available [11].

In the ozonation process, the static mixer is used to disperse the gaseous ozone into the liquid water stream for disinfection purposes.

2.1. Advantages of static mixers

Thakur et al. [11] have listed some advantages of static mixers over mechanically agitated tanks in the following table 1.

Table1: Advantage of static mixers over the mechanically agitated tanks [11].

Static mixer	Stirred tank reactor
Approach plug flow	The residence time is exponentially distributed
Small space needed	Large space needed
No power required except for pumping	High power dissipation
Self-cleaning	Large vessels to be cleaned
Small flanges to seal	Small flanges but with one large flange to seal
Good mixing at low share rates	Sensitive materials can be damaged at high shear rates
Fast product grade changes	Product grade changes might create waste

2.2. Types of static mixers

Static mixer is standard equipment in the process industry with many different types such as Kenics, Koch, Sulzer SMX and Ross LPD as shown in Figure 6.

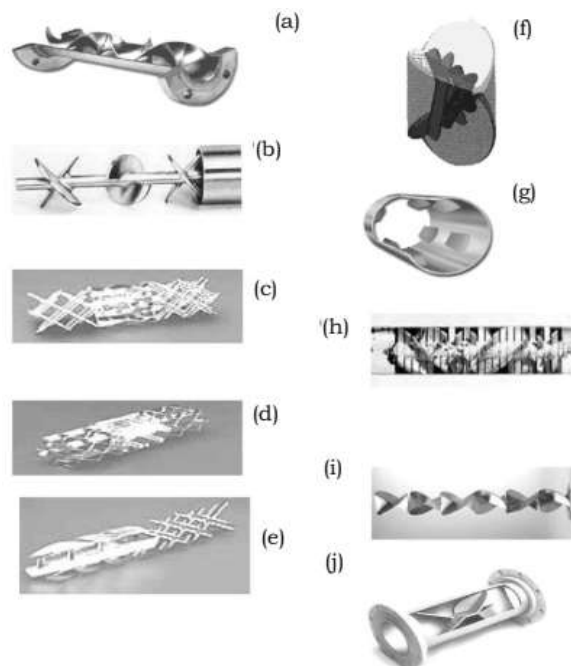


Fig. 6: (a) Kenics (Chemineer Inc.); (b) low pressure drop (Ross Engineering Inc.); (c) SMV (Koch-Glitsch Inc.); (d) SMX (Koch-Glitsch Inc.); (e) SMXL (Koch-Glitsch Inc.); (f) Interfacial Surface Generator-ISG (Ross Engineering Inc.); (g) HEV (Chemineer Inc.); (h) Inliner series 50 (Lightnin Inc.); (i) Inliner series45 (Lightnin Inc.); (j) Custody transfer mixer (Komax systems Inc.) [11].

The type of the static mixer used has great influence on the mixing mechanism, which, in turn, has a significant impact on the pressure drop and flow patterns of the fluids. Because of the light structure of the Kenics and Ross LPD, they can be applied for low viscosity fluids in either transient or turbulent flow conditions at low pressure drop [11, 23].

2.3. Kenics static mixer (KM)

The Kenics static mixer consists of a series of fixed helical elements or blades installed inside tubular housing as shown in Figure 7. KM provides continuous blending and dispersion of the flowing materials, with no moving parts, and no external power or regular maintenance, by redirecting the flow patterns present in the open pipe. The KM is used for liquid-liquid, solid-gas, gas-gas and gas-liquid mixing. When two immiscible fluids are exposed to shear forces inside the Kenics mixer, one of the phases will be dispersed in the other phase as droplets. The size of the drops depends on many parameters: the diameter of the mixer, velocity, elements shape and the physical characteristics of the both fluids. Small bubble size enhances the interfacial area between the two fluids, leading to higher mass transfer coefficient and therefore higher mass transfer between the two fluids. The Kenics static mixer has an advantage over other types of static mixer in that; it enhances the rate of mass transfer without wasting energy or material blockage. Moreover, the helical elements promote plug flow in continuous processes. The pressure drop increases along the mixer providing the energy need for mixing process [19, 11].



Fig. 7: The structure of Kenics static mixer [10]

The geometric structure of the helical elements produces the mechanism of simultaneous flow division and radial mixing. In the case of laminar flow, the flowing material is divided at the leading edge of each element and follows the channels produced by the element structure. The two channels produced by the first element are further divided by all the succeeding elements, leading to continuous increase in stratification as indicated in Figures 8 and 9. The number of stratification layers can be calculated by 2^n , where n is the element number [19].

The radial mixing in either laminar or turbulent flow is the rotational circulation of the flowing materials around their hydraulic centre in every single channel of the static mixer resulting in continuous and perfect mixing between the two flowing fluids. This mechanism eliminates the radial gradient in velocity, fluid composition and temperature and therefore reducing fouling and thermal degradation [19].

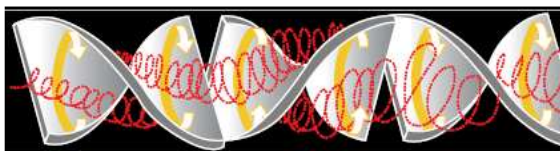


Fig. 8: The mixing mechanism inside a Kenics static mixer [10]

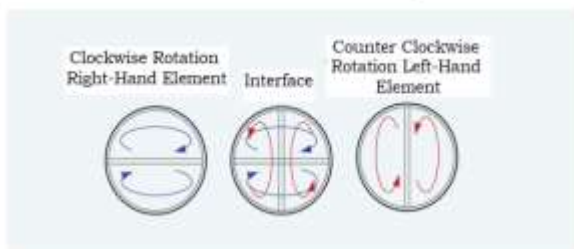
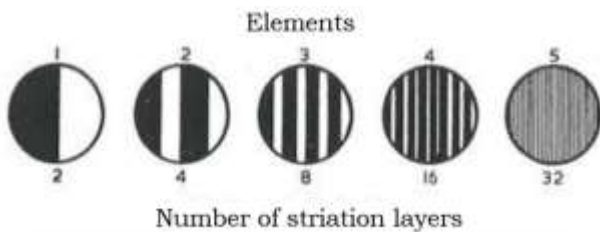


Fig. 9: Principles of operation of static mixer modules [19]

2.4. Application of static mixers in the process industry

Static mixers have been used in many fields of the chemical industry as shown in table 2.

Table 2: Application of static mixers in the chemical process industry adapted from [11, 19].

Industry	Applications
Chemical and agricultural chemicals	Chlorination, Reaction enhancement Gas mixing, Organic-aqueous liquid-liquid

Food processing	Fertilizer and pesticide preparation Liquid blending and emulsification Starch slurry and cooking Heating and cooling of sugar solutions Solid ingredient bending
Minerals processing	Mineral recovery by solvent extraction Slurry dilution, Oxidation and bleaching Chemical addition and bleaching
Petrochemicals and refining	Gaseous reactant blending Gasoline blending, Caustic scrubbing of H ₂ S and CO ₂ Lube oil blending
Pharmaceuticals and cosmetics	Mixing of trace elements Blending of multi-component drugs Dispersion of oils, Sterilization, pH control
Polymer, plastics and textiles	Continuous production of polystyrene Mixing of polymer additives, Preheating polymers, thermal homogenization
Water and waste water treatment	Addition of coagulating agent Disinfection (Cl ₂ , O ₂ , O ₃) Dechlorinating, pH control

3. Theory of Backmixing in gas-liquid contactors

It well established that backmixing has an important effect on the performance of the continuous fluid flow gas-liquid reactors. Backmixing is an intermediate flow pattern which falls between the ideal perfect mixing and plug flow [16]. There are two main non-ideal flow patterns as shown if Figure 10, channelling flow where the fluid elements flow as independent paths across the column and can be developed to back flow when the channels bend back on themselves. The second one is backflow where the fluid elements travel at velocities larger than the total average velocity. This is due to the fact that large back velocity will significantly reduce the forward velocity [16].

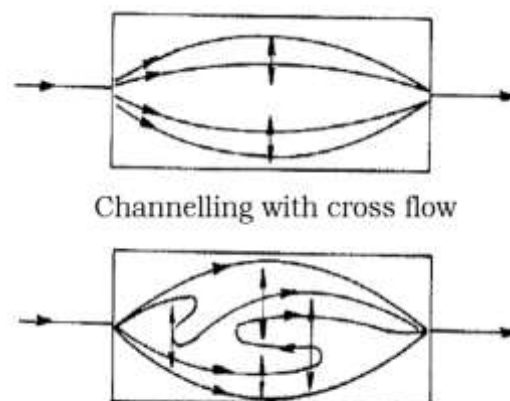


Fig. 10: Desegregated flow [16]

Generally, gas-liquid contactors have blades or baffles, which separate the channels because of transverse mixing so as to enhance the gas-liquid contact and create uniform conditions. In the case

of complete transverse mixing, channel flow turns to plug regime. When the amount of backflow is smaller than that of transverse mixing, backflow will approach the plug flow regime. However, if it is much larger than transverse mixing, the backflow will tend to perfect mixing regime. There is another important case which occurs when the transverse mixing and backflow are infinite with the same rate. This case is called backmixing where the flow regime is intermediate between the perfect mixing and plug flow [15, 16]. Two main models have been developed to study the effect of backmixing:

3.1. The stage wise backmixing model

The model shown in Figure 11 describes the backflow of fluid between the stages of the column in transverse direction to the main flow. It is assumed that the stages are completely mixed therefore fluid's elements can move forward or backward in case of neglecting the convective flow. The back mixing is the ratio of the backward flow between the cells or stages to the total convective forward flow which is equal to the exit of feed flow rate. This model applies to stage wise apparatus such as plate towers where the concentration profile changes in a series of steps [16].

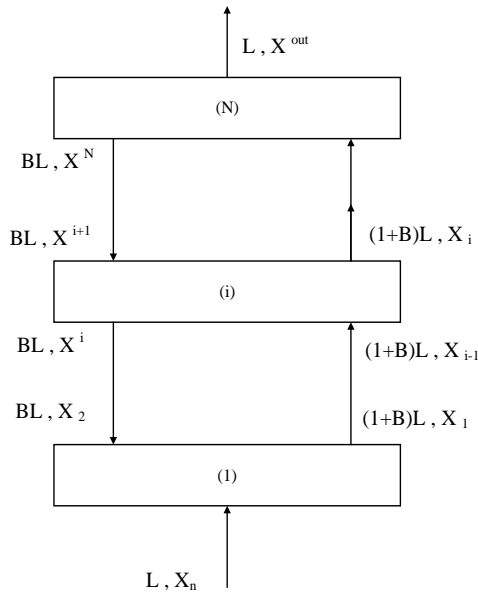


Fig. 11: Schematic representation of the stagewise backmixing model (adapted from [16]).

3.2. The differential backmixing model

In this model, backmixing can be represented as infinite transverse mixing and infinite backflow with same order as shown in Figure 12. Elements of fluid can travel forwards and backwards in relative to the main convective flow which can move in either direction. Thus, it can be represented by diffusion and Fick's Law:

$$uX = ux - D \frac{dx}{dl} \tag{12}$$

Where: U: is the fluid velocity, l is the flow per unit are, x is the concentration, ux: is the convective flow, (-Ddx/dl) is the diffusion flow and D is the eddy diffusion coefficient [16].

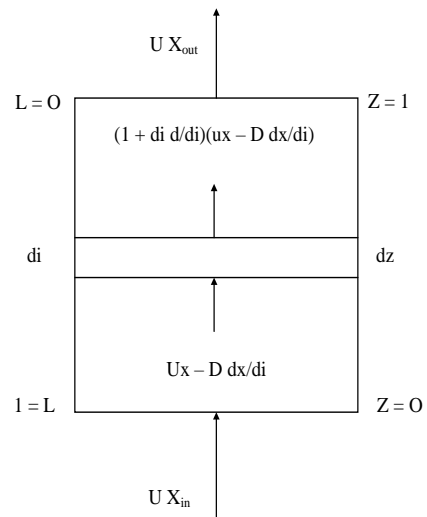


Fig. 12: Schematic representation of the differential backmixing model [16]

4. Residence time distribution (RTD) in flow reactors

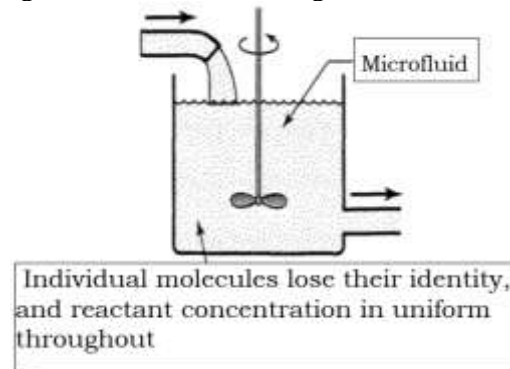
The residence time distribution (RTD) is a crucial factor for the study of the continuous chemical engineering processes occurring in a static mixer. It is the time that a fluid spends inside a static mixer. The RTD describes the degree of mixing, product's quality, and chemical reaction yields [15].

4.1. Non-ideal flow

Real reactors deviate from ideal reactors due to several factors: channelling, stagnant regions, short circuiting and backflow. For ideal continuous stirred tank reactors (CSTR), it is assumed that the concentration of the reactant is the same at any place inside the reactor. In real stirred tanks, however, the reactant concentration at the feed entrance is higher than its concentration in the stagnant regions and behind the impellers. In an ideal flow reactor, at any axial position, all the molecules of both reactant and product travel at the same rate in the direction of the bulk flow. Whereas, in the real one, because of molecular diffusion, turbulence mixing and velocity profile of the fluid, the molecules of the fluid travel at non-uniform velocity and different direction [15, 18]. Three factors can be used to account for this deviation:

Stage of aggregation

The state of aggregation depends on the nature of the flowing material. The fluid mixing in any phase at nonideal flow conditions is classified as shown in Figure 13 into: micromixing and macromixing.



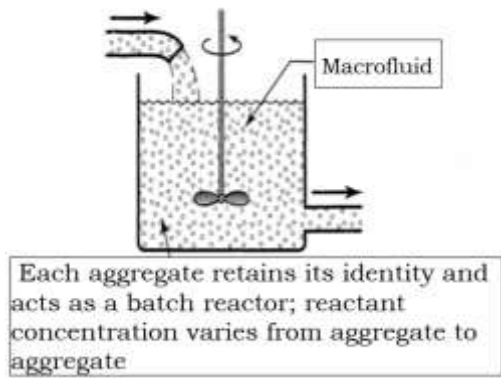


Fig. 13: Two extremes of aggregation of fluid [18]

Earliness of mixing

This factor is very important for the systems which have two input feed stream as shown in the Figure 14.

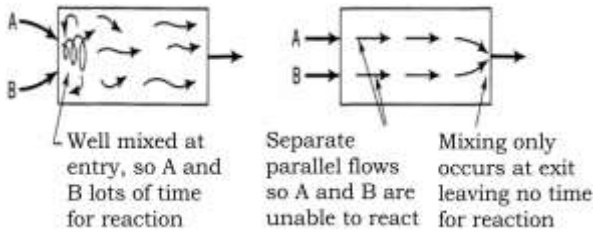


Fig. 14: The effect of early or late mixing on the performance of the reactor [18]

The age distribution of fluid (RTD or E)

Inside the reactor, fluid elements travel through different paths. Thus, they may spend a different periods of time to pass the reactor. The exit age distribution, E is defined as the distribution of those times of fluid stream exiting the reactor. The unit of the RTD is time^{-1} . RTD can be represented by assuming that the area under the curve is equal to unity as shown in Figure 15.

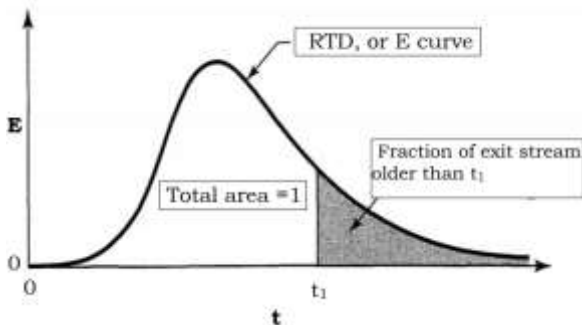


Fig. 15: The residence time distribution of fluid flowing through a contactor [18]

4.2. Methods for measuring residence time distribution (RTD)

The characteristic of backmixing of different phases in gas-liquid reactor are determined from the residence time distribution of a tracer which is fed to the system [15] The most common methods that can be applied to determine the RTD are an impulse, step-change input of the tracer as shown in the Figure 16.

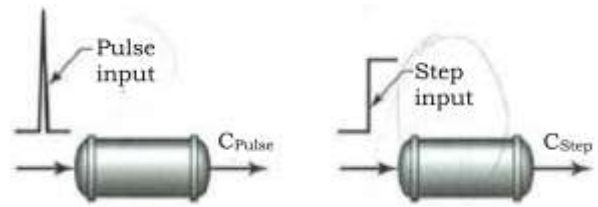


Fig. 16: The pulse and step tracer [18]

Impulse tracer

In this method, a small amount of tracer is injected at the entrance of the reactor during a period of time. This time period is much shorter than the mean residence time. The outlet concentration of the tracer is determined as function of time [18,19].

Step change

If the feed stream to the reactor is replaced instantaneously with another one, the step change experiment can be conducted. Both fluids should have the same behaviour within the system and also have the same viscosity and density [19].

4.3. The transient backflow cell model (TBFCM)

The developed models for designing gas-liquid reactors must describe the flow and mixing conditions inside the reactor. The most common ideal reactors are that used to design ozone contactor are the plug flow reactor (PFR) and the continuous flow stirred tank reactor (CFSTR). The PFR approaches the plug flow conditions. Therefore, the mixing between the adjacent flow cells is not permitted whereas, the CFSTR is considered to be perfectly mixed and has uniform concentration along the column [18]. Due to backflow, short-circuiting and stagnant zone, these two ideal flows are no longer applicable to describe the real flow inside the ozone contactors. The residence time distribution can be used to analyse these complicated flow characteristic in the ozone contactor [15, 26].

The ordinary stirred tanks in series model which assume perfect mixed cells has been employed to describe the mixing process. However, this model does not take into consideration the upstream mixing of material i.e. the mixing in direction opposite to the direction of the main flow. In order to overcome this problem, the backflow cell model has been developed by Mecklenburg and Hartland [16]. The BFCM introduces the backflow between the cells inside the gas-liquid contactor. The backflow ratio, B is the main parameter in the BFCM. The performance of the back flow cell model varies according to the value of the backflow ratio from ordinary tanks in series ($B \rightarrow 0$) to single stirred tank ($B \rightarrow \infty$) [2,27-29,33].

The BFCM is a mathematical model that is applied to characterise the performance of the Kenics ozone static mixer. In order to describe the axial dispersion in the liquid phase, the BFCM assumes a back flow between the cells in direction opposite to the main liquid flow and exchange flow in the same direction of the main liquid flow. These two flows have been expressed in our study as back flow ratio (B) and exchange flow ratio (B) and both of them are assumed to be equal and constant along the mixer. Generally, BFCM is composed of two series of equal number of completely mixed cells in which one series describe the liquid phase

and the other describes the gas phase [17]. In this model, the backmixing in the gas phase was assumed to be negligible because of the large buoyancy of the gas bubbles, gas and liquid flow rates, interfacial and gas hold-up are constant along the contactor. Roemer and Durbin [28] have developed very efficient TBFCM to describe the residence time distribution inside the chemical reactors as shown in the Figure 17 below.

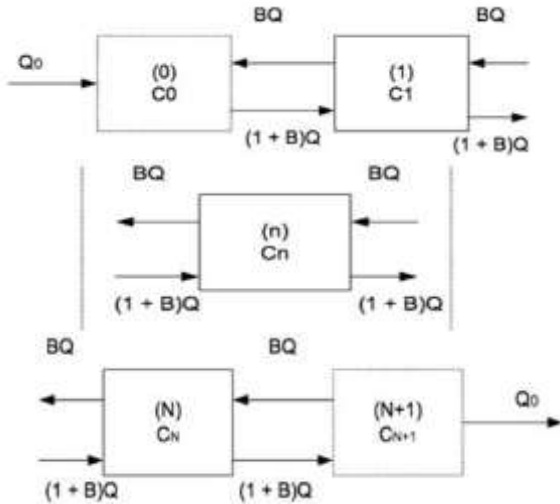


Fig. 17: The transient backflow cell model [28]

This model consists of number of completely mixed cells (N) that have an equal volume ($V_c = V_T/N$) with equal backflow rates between the cells. The cells: (0 and N+1) have negligible volume or hold-up and they allow the boundary conditions to be easily determined.

The material balance has been carried out around each cell with respect to the inert tracer and the following equations are produced:

$$n = 0, \quad \delta(t) = (1 + B)E_0 - Bg_1 \quad (13)$$

$$1 \leq n \leq N, \quad \frac{1}{N} \frac{dE_n}{dt} = (1 + B)E_{n-1} - (1 + 2B)E_n + BE_{n+1} \quad (14)$$

$$n = N, \quad 0 = E_{N-1} - E_N \quad (15)$$

These equations have been transformed to the following equations:

$$E(\theta) = \sum_{i=1}^N \{A_i \exp(s_i \theta)\} \quad (16)$$

Where: $E(\theta)$ is the impulse response of N^{th} cell at time θ , ($C_i(\theta)/C_i^*$)

For $0 < B < \infty$ or $0 < \lambda < 1$, the distinct poles of the transfer function can be calculated as:

$$s_i = \left(\frac{N}{1 - \lambda} \right) [2\lambda^{0.5} \cos(\vartheta_i) - (1 + \lambda)] \quad (17)$$

With $1 \leq i \leq N$

$$A_i = (-2N\lambda^{-N/2})(\sin^2(\vartheta_i)/D'(\vartheta_i)) \quad (18)$$

$D'(\vartheta_i)$ is the derivative of the function $D(\vartheta_i)$ which is equal to:

$$D(\vartheta) = \lambda^{0.5} \sin((N + 1)\vartheta) - 2 \sin(N\vartheta) + \lambda^{0.5} \sin(N - 1)\vartheta \quad (19)$$

Where ϑ_i are the roots of the equation $D(\vartheta) = 0$ in the interval $0 < \vartheta_i < \pi$

The above equations have been solved in order to produce the impulse residence time distribution of the ozone solute.

Before measuring the impulse response, the roots, ϑ_i of the function $D(\vartheta)$ have been determined by the Newton-Raphson method. Newton method is modified Taylor series method and uses iterative techniques to solve the non-linear algebraic equation $D(\vartheta) = 0$. Initial guess of the (ϑ_i) has to be made in the interval $(0 < \vartheta_i < \pi)$. Newton method is very fast and efficient and it has the following general formula:

$$\vartheta(i + 1) = \vartheta(i) + \frac{D(\vartheta_i)}{D'(\vartheta_i)} \quad (20)$$

The iteration stops when the function $D(\vartheta)$ value satisfies the following conditions:

$D(\vartheta_i) \leq \delta_{abs}$ (tolerance) [30]. It should be noted that the number of the roots, ϑ_i of the function $D(\vartheta)$ is equal to the number of cells used and their values depends on the cell number and the back flow ratio which is expressed in this model by the term (λ).

Gamal El-Din and Smith, [31] have developed a transient BFCM as alternative approach to the axial dispersion model and continuous flow stirred tanks in series in order to describe the hydrodynamics and the back mixing in the liquid phase in the ozone bubble contactor. A schematic representation of their model is shown in Figure 18.

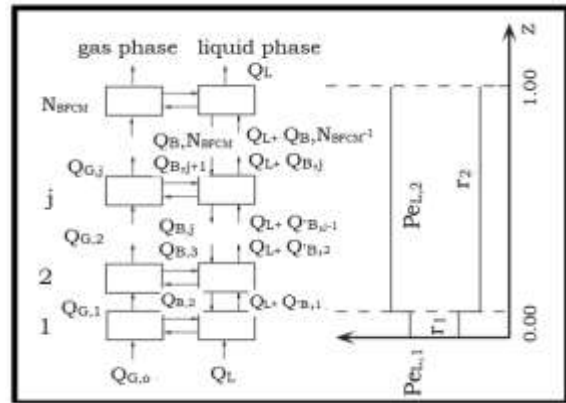


Fig. 18: Schematic diagram representing the co-current transient back flow cell model [31]

In their study, they have studied the effect of number of cells (N_{BFCM}) and the Peclet number (Pe_L) on the theoretical RTD curves as shown in the Figures 19 and 20.

They found that as the N_{BFCM} and Pe_L increase, the RTD curves become narrower, more symmetrical and higher peak. They have ascribed this to the fact that the liquid flow approaches the plug flow pattern at high values of Peclet number [33].

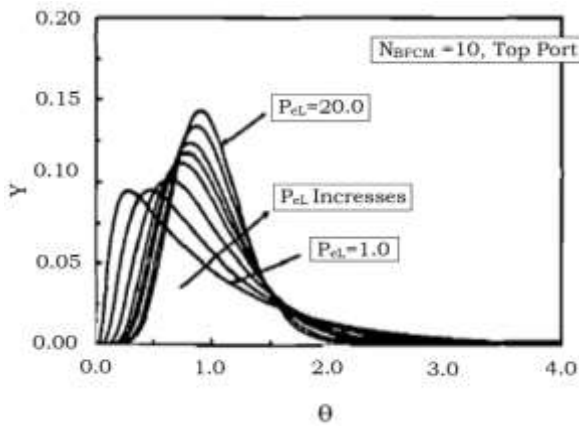


Fig. 19: Theoretical RTD curves for different Peclet numbers [31]

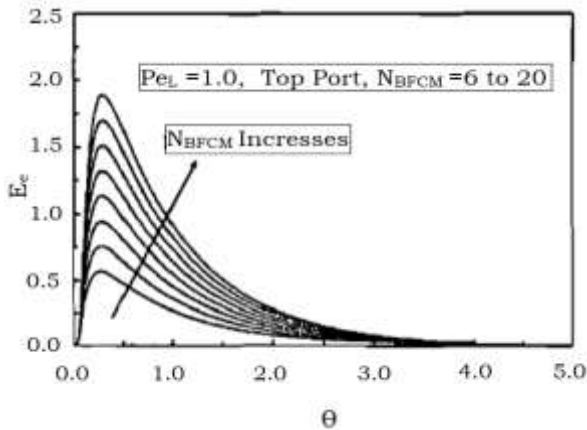


Fig. 20: Effect of the number of cells on the RTD curves [31]

As shown in the Figures 21 and 22 below, the RTD predictions the three models: TBFCM, ADM and CFSTR's in series have been validated with RTD experimental data of Zhou, 1995 [8]. They concluded that the TBFCM was better than other models in characterising the backmixing between the cells.

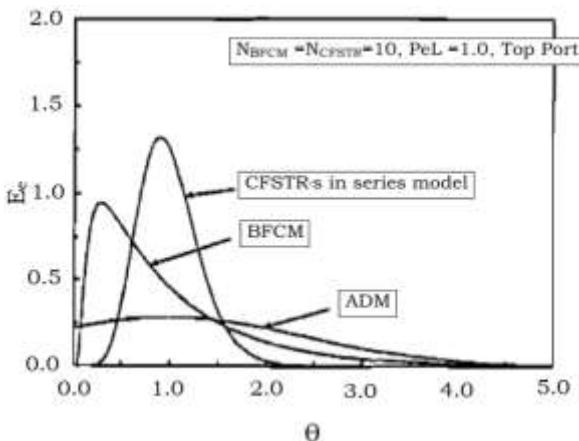


Fig. 21: Comparison between the TBFCM, ADM and CFSTR's models [31]

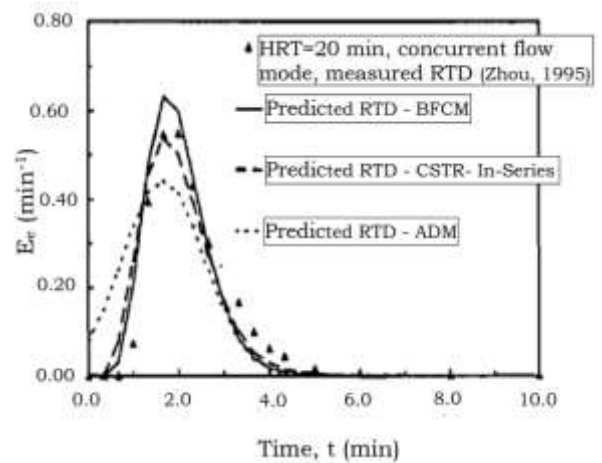


Fig. 22: Comparison between the TBFCM, ADM and CFSTR's in estimating the RTD curve for co-current bubble column [31]

Baawain et al. [24] have also developed TBFCM to characterise the hydrodynamic of co-current impinging-jet ozone bubble column. They have carried out tracer test on the bubble column which had inside diameter of 100 mm and height of 1520 mm in order to study the hydrodynamic of the column. The superficial gas velocity range was from 0.002 to 0.014 m s⁻¹ and the range of the superficial liquid velocity was from 0.008 to 0.028 m s⁻¹.

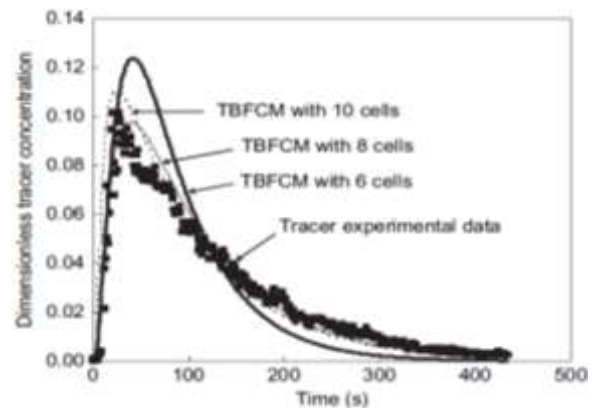


Fig. 23: Effect of the number of cells of TBFCM on the RTD predictions [24]

As shown in Figure 23, the TBFCM has been run at three different cell numbers in order to determine the most appropriate cell number that accurately describe the backmixing in the liquid phase. They found that as shown in figure that the TBFCM with 8 cells is best fitted with experimental data and therefore the most appropriate cell number.

5. Mathematical Model

The back flow cell model has been developed by Mecklenburgh and Hartland [16] as alternative approach to solve the equations of traditional axial dispersion model (ADM). The BFCM is a finite difference approximation of the ADM. In BFCM, the gas-liquid contactor length is represented by a number of completely mixed cells, which are connected to each other by the main flow of the gas and liquid phases, and the recirculation between the cells which depends on the axial dispersion inside the contactor.

5.1. Axial dispersion model (ADM)

This model simply characterises the backmixing by one-dimensional and diffusion process superimposed on plug flow equation which is expressed by the Fick’s law [24] The ADM uses only one parameter (Peclet number) to characterise the back mixing. Thus, ADM became simple and widely used model [15]. If an ideal tracer is injected to the reactor, it will spread as it travels through the column. The dispersion coefficient D (m^2/s) exemplifies the spreading process. According to the dispersion coefficient value, we have three cases: firstly, when D is equal to zero, no spreading, thus plug flow. Secondly, small D results in slow spreading of the tracer curve. Thirdly, large D results in rapid spreading [18]. The dispersion coefficient can be represented by the dimensionless Peclet number ($Pe = D/uL$) in order to characterise the spreading in whole reactor, Pe is used to define the degree of backmixing. When $Pe = 0$, we have complete backmixing, and if $Pe = \infty$, plug flow exists [15]. For the ADM, the residence time distribution (E_θ) can be expressed by [18].

$$E_\theta = \frac{1}{2\sqrt{\pi/Pe}} \exp\left(-\frac{Pe(1-\theta)^2}{4}\right) \quad (21)$$

5.2. Continuous stirred tank in series model (CSTR’s in series)

It is the simplest among stagewise models for characterisation backmixing in the multiphase reactors. In the CSTR’s in series model, the reactor is viewed as a series of completely mixed stages. The degree of backmixing is determined by stage’s number. The smaller the number of stages, the more significant is the backmixing [15]. Generally, CSTR’s in series model is more reliable than the ADM at high values of the dispersion coefficient [25]. The residence time distribution (E_θ) can be determined by [19].

$$E_\theta = \frac{N_{CFSTR} (N_{CFSTR} \theta)^{N_{CFSTR} - 1}}{(N_{CFSTR} - 1)!} \quad (22)$$

5.3. Relationship between the ADM and the transient BFCM

The transient BFCM is easier to formulate and solve than the ADM. This is because the BFCM produces non-linear algebraic equations whereas; the ADM produces non-linear differential equations which have to be converted to non-linear algebraic equations.

Both transient BFCM and ADM characterise the backmixing of the liquid phase but in different ways. BFCM describes the backmixing by using the backflow ratio (B) and number of the cells (N_{BFCM}) whereas the backmixing in the ADM is characterised by the dimensionless Peclet number. The back flow ratio (B) and Peclet number (Pe) can be interrelated by the following equation:

$$B = \frac{N_{BFCM}}{Pe_L} - 0.5 = \frac{D_L \epsilon_L N_{BFCM}}{u_L L} - 0.5 \quad (23)$$

$$\lambda = \frac{B}{1+B} \quad (24)$$

Where: N_{BFCM} : cells number, B = Back flow ratio = exchange flow ratio, D_L = axial dispersion coefficient in the liquid phase (m^2/s), u_L = superficial liquid velocity, ϵ_L liquid phase hold-up [2, 17, 24, 32].

5.4. Steady state back flow cell model (BFCM)

The steady state back flow cell model has similar characteristic to the TBFCM in that it can predict the hydrodynamic of the ozone contactor with variable back mixing, variable cell volume or cross sectional area and pressure along the column [24]. The assumptions that generally govern the steady state BFCM is similar to the TBFCM. In addition to those assumptions: (1) the resistance to the mass transfer is limited only to the liquid phase; (2) slow chemical reaction, therefore the enhancement factor is equal to 1; (3) the process is operated at steady state conditions; (4) Henry’s law applies for the equilibrium concentration of ozone in water; (5) the rate of ozone auto-decomposition is described by a pseudo first order rate expression in the liquid phase [25].

Gamal El-Din and Smith [25] have developed steady state BFCM in order to measure the ozone concentration profile in co-current and counter-current bubble column. In their model, they assumed variable backmixing, variable cross-sectional area and variable cell volume across the column. They performed mass balance with respect to ozone solute around each cell inside the column. The following equations were produced, for $j = 1, 2 \leq j \leq N_{BFCM}$ and $j = N_{BFCM}$ respectively.

$$Q_L Q_{L,0} + Q_B Q_{L,2} + k_L a (C_{L,1}^* - C_{L,1}) \frac{V}{N_{BFCM}} - k_w C_{L,1} \epsilon_L \frac{V}{N_{BFCM}} - (Q_L + Q_B) C_{L,1} = 0 \quad (25)$$

$$(Q_L + Q_B) C_{L,j-1} + Q_B C_{L,j+1} + k_L a (C_{L,j}^* - C_{L,j}) \frac{V}{N_{BFCM}} - k_w C_{L,j} \epsilon_L \frac{V}{N_{BFCM}} - (Q_L + 2Q_B) C_{L,j} = 0 \quad (26)$$

$$(Q_L + Q_B) C_{L,N_{BFCM}-1} + k_L a (C_{L,N_{BFCM}}^* - C_{L,N_{BFCM}}) \frac{V}{N_{BFCM}} - k_w C_{L,N_{BFCM}} \epsilon_L \frac{V}{N_{BFCM}} - (Q_L + 2Q_B) C_{L,N_{BFCM}} = 0 \quad (27)$$

Their mass balance equation of gaseous ozone in the gas phase for cell j is:

$$Q_{G,j-1} Q_{G,j-1} - Q_{G,j} Q_{G,j} - k_L a (C_{L,j}^* - C_{L,j}) \frac{V}{N_{BFCM}} = 0 \quad (28)$$

By rearranging the equations above, the following steady state BFCM equations are produced:

Ozone solute in the liquid phase for $j = 1, 2 \leq j \leq N_{BFCM}$ and $j = N_{BFCM}$ respectively.

$$X_0 - (1 + r + D_a + St_L) X_1 + r X_2 + St_L (1 + \alpha/2) Y_1 = 0 \quad (29)$$

$$(1 + r) X_{j-1} - (1 + 2r + D_a + St_L) X_j + r X_{j+1} + St_L [1 + \alpha(j - 0.5)] Y_j = 0 \quad (30)$$

$$(1 + r) X_{N_{BFCM}-1} - (1 + r + D_a + St_L) X_{N_{BFCM}} + St_L [1 + \alpha(N_{BFCM} - 0.5)] Y_{N_{BFCM}} = 0 \quad (31)$$

Ozone solute in the gas phase, for cell j :

$$(1 + \alpha(j - 1.5)) q_{G,j-1} Y_{j-1} - (1 + \alpha(j - 0.5)) q_{G,j} Y_j - St_{LG} [(1 + \alpha(j - 0.5)) Y_j - X_j] = 0 \quad (32)$$

Where: $r = Q_B/Q_L$, $St_L = k_L a/N_{BFCM} u_L$, $St_G = k_L a L R T / N_{BFCM} u_{G,0} H$, $D_a = k_w \epsilon_L L / N_{BFCM} u_L$, $X_j = C_{L,j} / C_{L,0}^*$, $Y_j = y_j / y_0$ and $q_{G,j} = Q_{G,j} / Q_{G,0}$

These equations have been solved using TK Solver software at number of cells, ozone decay rate and tolerance of 10, 0.028 min⁻¹, and 10⁻⁶ respectively.

They validated their model with the experimental results of Zhou [7], where bubble column with diameter of 100 mm and a height of 2000 mm were used. The predicted ozone concentration profile along the column is shown in Figure 24.

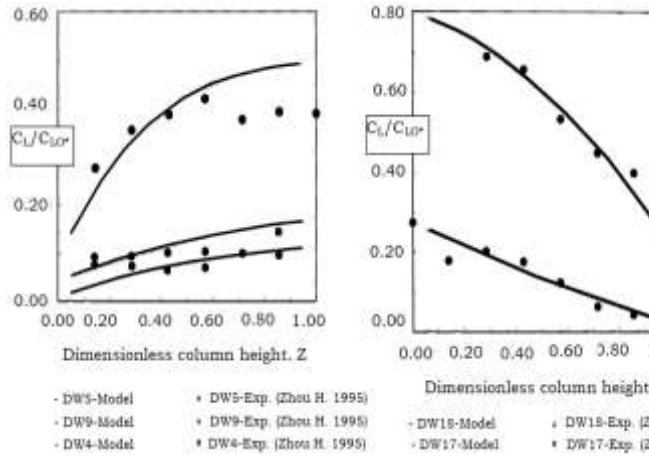


Fig. 24: (a) co-current steady state BFCM and (b) counter-current steady state BFCM [8]

Baawain et al. [24] have modified the steady state BFCM mentioned above in order to account for variable mass transfer coefficient along the height of the impinging-jet bubble column. This assumption is described by the varying liquid phase Stanton number (St_L). Also, the variables cell volume, cross-sectional area and pressure has been described by the following parameters: f_j, f_{A,j} and f_{Z,j} respectively.

For cell: j = 1

$$X_0 - (1 + r_1 + f_j \cdot f_{A,j} \cdot D_{A,j} + f_j \cdot f_{A,j} \cdot St_{L,j}) \cdot X_1 + r_1 \cdot X_2 + f_j \cdot f_{A,j} \cdot St_{L,j} \cdot (1 + \alpha \cdot f_{Z,j} \cdot 0.5) \cdot Y_1 = 0 \quad (33)$$

For cells: 2 ≤ j ≤ N_{BFCM}-1

$$(1 + r_{j-1}) \cdot X_{j-1} - (1 + r'_j + r_j + f_j \cdot f_{A,j} \cdot D_{A,j} + f_j \cdot f_{A,j} \cdot St_{L,j}) \cdot X_j + r'_{j+1} \cdot X_{j+1} + f_j \cdot f_{A,j} \cdot St_{L,j} \cdot (1 + \alpha \cdot f_{Z,j} \cdot (j - 0.5)) \cdot Y_j = 0 \quad (34)$$

For cell: j = N_{BFCM}

$$(1 + r_{N_{BFCM}}) \cdot X_{N_{BFCM}} - (1 + r'_{N_{BFCM}} + f_j \cdot f_{A,j} \cdot D_{A,N_{BFCM}} + f_j \cdot f_{A,j} \cdot St_{L,N_{BFCM}}) \cdot X_{N_{BFCM}} + f_j \cdot f_{A,j} \cdot St_{L,N_{BFCM}} \cdot (1 + \alpha \cdot f_{Z,j} \cdot (N_{BFCM} - 0.5)) \cdot Y_{N_{BFCM}} = 0 \quad (35)$$

For the ozone solute in the gas phase:

For cells: 1 ≤ j ≤ N_{BFCM}

$$q_{G,j-1} \cdot Y_{j-1} \cdot (1 + \alpha \cdot f_{Z,j} \cdot (j - 1.5)) - q_{G,j} \cdot Y_j (1 + \alpha \cdot f_{Z,j} \cdot (j - 0.5)) - f_j \cdot f_{A,j} \cdot St_{G,j} \cdot ((1 + \alpha \cdot f_{Z,j} \cdot (j - 1.5))) \cdot Y_j - X_j = 0 \quad (36)$$

TK Solver software and Newton method were used to solve the above mathematical equations with ozone decay rate of 2.61 × 10⁻⁴ s⁻¹. Their model

predicted the dissolved ozone concentration profile along the column as shown in Figure 25 below.

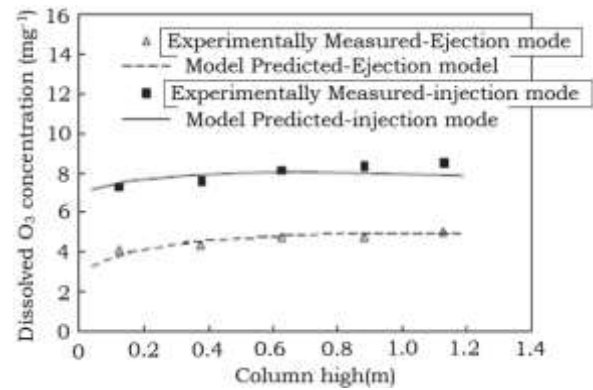


Fig. 25: Dissolved ozone concentration profiles for injection and ejection methods [24]

It was concluded that the model accurately predicted the concentration profile of the dissolved ozone. Tizaoui and Zhang [2] have developed steady state BFCM to describe mass transfer of ozone from the gas phase to the water inside Kenics static mixer. The volumetric mass transfer coefficient has been used to describe the liquid phase resistance to the ozone mass transfer.

The steady state BFCM results have been validated with experimental data. The experiment was carried out by using two static mixers of different lengths (0.37 and 0.74m) at gas flow rates range (400, 600, 800 mL/min) and liquid flow rates (1.5, 1.8, 2.3 L/min) and different entrance gas concentrations (64 and 82 g/m³ NTP).

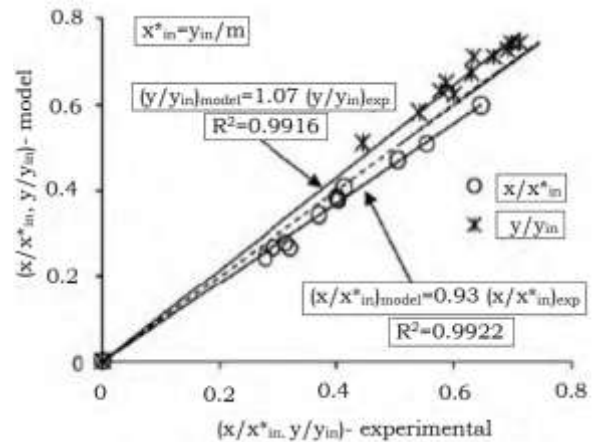


Fig. 26. The relation between model and experimental outcome [9]

As shown in Figure 26, the model was verified comparing the experimental and predicted values of outlet ozone concentration in the gas and liquid phases and good agreement between experimental and calculated values was obtained.

6. Conclusion

This paper has studied the use of static mixers for water ozonation, Application of static mixer in other industries, types and advantages, focusing on a ozone mass transfer in Kenics static mixer. Moreover, the basic knowledge and techniques that are needed for developing the mathematical models are discussed. Different mathematical models: BFCM, ADM and CFSTR have been discussed in this paper.

Gas-liquid contactors produce high ozone concentration. However, they require low gas flow rate that cause low gas to liquid volumetric flow rate ratio. However, static mixers are more efficient at low gas/liquid ratio and high ozone concentration. Furthermore, they produce a homogeneous ozone concentration because of the high turbulence produced by the elements that are inside the mixer [1,2].

As a result of this review, the steady state BFCM was verified by comparing the experiment data of Tizaoui and Zhang [2] and calculated-from-the-model values of ozone concentration in the gas and liquid phases at the outlet of the Kenics static mixer [2]. A satisfactory correlation between experimental and calculated values was obtained with an error of about 5%[2]. Furthermore, both BFCM and ADM have proved to be accurate, sufficient and reliable in their predictions of the performance of the ozone Kenics static mixer. However, the transient BFCM have provided slightly better results than the ADM. This is because the transient BFCM uses two mixing parameters to characterise the backmixing in the liquid phase: the number of cells and the backmixing ratio whereas the ADM uses only one parameter which is the Peclet number.

7. Reference

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