CHAPTER 1

INTRODUCTION

1.1 MOTIVATION FOR RESEARCH WORK

New technologies in chemical process industries are extending the usage of particulate solids in the mixture state. Mixing of powders, particles, flakes, fibres and granules gain increasing economic importance in different types of industry ranging from mixing of human and animal foodstuff, pharmaceutical products, detergents, chemicals and plastics etc.

The term mixing is applied to operations which tend to reduce non-uniformities or gradients in composition, properties, or temperature of material in bulk. Such mixing is accomplished by movement of material between various parts of the whole mass (Weinekotter et al 2000).

The most important use of mixing is production of homogenous blend of several ingredients that neutralizes difference in concentration inside the volume of a batch. However, mixing processes are not always designed with appropriate care. This causes significant financial loss which arises in two ways: i.e. either a) Mixture quality is poor; or b) The homogeneity is satisfactory but too much energy is consumed in achieving it. Hence both design and operation of the mixing unit itself will have a strong influence on the quality produced. However, research on these problems by analyzing different materials and conditions for achieving necessary quality by means of direct empirical work is a long and expensive procedure. Hence, the need for mathematical models is growing up. The models can dramatically reduce the empirical work required for predicting mixture parameters. It results from the fact that a couple of simple experiments can be made to determine the parameters of a model and afterwards the model will give information about the process when mixing conditions change. Without the model, it would require many experiments with the equipment to be able to find the best regime or to investigate how this equipment reacts to the changes in mixing conditions. Thus, simulation results in cutting costs of empirical work and decreasing the time for experiments.

1.2 LITERATURE SURVEY

Due to the industrial importance of static mixers, many studies have been undertaken attempting to characterize their performance. This chapter provides a two-part review of previous investigations on static mixers. The first part reviews experimental studies on performance of static mixers. The second part provides information about the mathematical modeling for Residence time distribution.

1.2.1 Literature Survey on Static Mixers

Many experimental studies have been undertaken in an attempt to characterize the performance of static mixer in both laminar and turbulent flow regimes.

During the past few years several immobile in-line mixers have been introduced to the industries to achieve the intermixing of process streams. A list of commercial static mixers and their applications has been described by Pattison (1969). Piters et al (1973), Simpson (1974), Morris et al (1974). Pahl and Muschelknautz (1982) have compared 12 different static mixer types, but considering only global properties, like pressure drop and overall mixing efficiency. Arimond and Erwin (1985), Khakhar et.al. (1987), Muzzio et.al (1998), Hobbs and Muzzio (1997a,b), Hobbs et.al. (1998) numerically elucidated the velocity Belt, pressure drop, and residence time distribution of the static mixers. Most related works considered the aspect of macro mixing, by elucidating the residence time distribution (RTD) of mixers (Nigam and Vasudeva 1980, Nauman 1982, 1991, Nauman and Vasudeva 1980, Kemblowski and Pustelnik 1988). However, as Khakhar et al (1987) addressed, that RTD cannot properly reflect the complicated flow characteristics in the static mixers.

The use of static mixer as a chemical reactor has been described by Bor(1971). The steady state continuous plug flow reactor is one particular type of reactor having application in numerous industries, such as the manufacture of butadiene by the dehydration of butylenes. The design equation for this type of reactor is:

$$\frac{V_R}{F} = \int_0^X \frac{dX}{r} \tag{1.1}$$

where

V_R = Volume of reactor corresponding to a conversion (litre)

F = Mass feed rate gmmole/min

X = Conversion

r = Rate of reaction (gmmole/litre min)

The rate of reaction is affected by the degree of mixing of the reactants inside the reactor, in addition to the prevailing temperature, pressure and composition parameters.

But the volume of reactor can be written as

$$V_R = TN_T\chi \tag{1.2}$$

where

T = Residence time required in the reactor (seconds),

N_T = Molar flow rate gmmole/sec.

χ = Molar volume (litre/gm-mole)

Hence equation (1.1) can be written as

$$\frac{T}{F} = \int_0^X \frac{dX}{N_T \chi(r)} \tag{1.3}$$

The velocity profile for the flow of fluids, under laminar conditions through an annulus is given by:

$$V(r) = \frac{\Delta p}{4\omega L}\left[1 - \left(\frac{r}{R}\right)^2 + \frac{1-k^2}{\ln\left(\frac{1}{k}\right)}\ln\left(\frac{r}{R}\right)\right] \tag{1.4}$$

where

V(r) = Velocity of fluid element at a radial distance of ‘r’ (cm/sec),

ΔP = Pressure drop (gm/cm2)

R = Outer radius of annulus (cm),

k = Dimensionless number,

ω = Viscosity of fluid (gm/cm-sec),

L = Length of annulus (cm),

The average velocity is obtained by integrating equation (1.4) within the limits k to R and dividing by the area of annulus.

Hence the average velocity is given by

$$V_{ave} = \frac{\Delta P R^2}{8\omega L}\left[\frac{1-k^4}{1-k^2} - \frac{1-k^2}{\ln\left(\frac{1}{k}\right)}\right] \tag{1.5}$$

The maximum velocity occurs at a radial distance of λ_R which is given by

$$\lambda_R = \left[\frac{1-k^2}{2\ln\left(\frac{1}{k}\right)}\right]^{1/2} \tag{1.6}$$

It can be observed from each equation (1.4) and equation (1.6) that the profile is not simple parabolic and the flatness of velocity profile predominates very close to the point of maximum velocity.

Kundsen et al (1958) and Meter et al (1961) studied the turbulent flow in annulus. The experiments indicated that the maximum velocity in turbulent flow also occurred at a distance of λ_R defined by equation (1.6). The velocity profile is much more flattened throughout the cross section (Meter et al 1961) thus, indicating a plug flow condition in the annular space.

Helical coils are widely used in the process industries to improve the mixing efficiency under laminar flow conditions (Monisha Mridha et al 2007).

An attempt has been made to further enhance the mixing in the coiled tube at low Dean Number using the phenomenon of flow inversion. The study is performed in coiled flow inverter (CFI) by Saxena and Nigam (1984) with coiled configuration for flow inversion and its effect on residence time distribution. Monisha Mridha et al (2007) developed the concept of inverting the direction of fluid by 90º. It comprises coils with equidistant 90º bends.

The goal of the work is to measure mixing performance in the static mixer. Studies have characterized mixing performance via Residence Time Distributions (Wang et al 2008, Nauman et al 2002, David Olivet et al 2005). Characterization of static mixers is based either on the results obtained by experiment, or on an over-simplified model of the mixer behavior, without detailed measurement or calculation of the physical features of the flow. This sort of characterization represents a significant step forward toward understanding the actual performance of static mixers.

1.2.2 Literature Survey on Residence Time Distribution

Residence time distribution (RTD) of particles is one of the critical factors that play a role in aseptic processing. In a continuous flow system, some of the particles remain in the system longer than others, i.e. particles close to wall or in a dead space move much slower than those traveling along the centerline. Hence, the particles experience a distribution of residence times within the system. Residence time of particles must be as narrow as possible in order to reduce the degree of over processing and obtain a uniformly heated product (Ramaswamy et al 1995).

Residence time distributions can be described by E(t) and F(t) functions. The E(t) function gives the RTD of the fluid or particles for any non-ideal flow. A typical E(t) curve will describe the fraction of material in the outlet stream that has been in the system between times t and t+dt. The

F(t) function represents the accumulation of particles at the exit with a residence time t or less. F(t) can be obtained by integrating E (t) over time t (Ramaswamy et al 1995). Standard deviation of residence times has been used by researchers to characterize RTD and to determine the spread of the distribution.

1.2.2.1 Techniques to determine residence time

There are several studies which involve determination of residence time distribution of liquid or multiphase media. There are various types of methods used in determination of RTD. These include optical methods, use of magnetic particles, magnetic resonance imaging, chemical tracers, fractional collection, metal detectors, radioactive tracers, and ultrasonic Doppler velocimetry methods.

Introducing a tracer solution has been widely used in determining RTD of liquids. In a study by Bateson (1971), age distribution of fluid particles (water and starch solution) in a heat exchanger was determined by introducing 20% saline solution. Milton and Zahradnik (1973) performed their research for a heat-cold-cool system. A pulse type of 60% sucrose solution was injected as a tracer material in a polyethylene glycol solution which was the carrier fluid. The concentration of sucrose in the fluid leaving the system was analyzed by a polarimeter. Sancho and Rao (1992) used a pulse input of sodium chloride (NaCl) solution to determine RTD of water, sucrose solution, and guar gum solutions flowing in a straight holding tube.

Abichandini and Sarma (1988) performed a similar analysis to determine the RTD of water in a scraped surface heat exchanger. A pulse input of saturated NaCl solution was used as the disturbance at the inlet and the response was measured (at periodic intervals of 5 seconds) at the outlet by a conductivity bridge. Similarly, KCl and NaCl solutions and conductivity meter were used by Zhang et al (1990) and Lee et al (1995) to determine the RTD of fluids.

Dickerson et al (1968) used radioactive tracers to determine the residence time of milk products such as cream, ice cream mix, condensed skim milk, chocolate milk, and raw milk. Radioactive iodine, silver iodide crystals, and bacteria stained with iodine were different types of tracers used in this study. Torres et al (1998) used a dye, methylene blue, as a tracer in residence time experiments.

In an early study by Roig et al (1976), red dye was used to determine the RTD of water in the holding section of a plate heat exchanger. After introducing a step change in the concentration of the dye, samples at the outlet stream were collected at 2 seconds intervals and analyzed by a colorimeter at 625 and 430 nm. Taeymans et al (1986) also used dyes to determine RTD of liquids containing particles. For this purpose, tracer calcium alginate beads were colored in blue dye before entrapping spores in them.

Hall Effect sensors have been used by many researchers to investigate the RTD of particles in carrier fluids. Hall Effect sensors consist of a semiconductor device whose ability to pass electrical current is influenced by the presence of an external magnetic device. So, when a magnet moves through the sensing field of the sensor, the output current is detected. Furthermore, with a stop watch, residence time can be calculated if the distance between the two sensors is known (Tucker and Heydon 1998). This approach was used by Tucker and Withers (1994), Fairhurst et al (1999) and Eliot-Godereaux et al (2001). Although this method was shown to be promising, the primary drawback was its inapplicability to multiple particle systems.

Yang and Swartzel (1991) investigated the use of photo electronic sensors to determine the residence time distribution of particles in a continuous thermal processing system.

Chandarana and Unverferth (1996) used magnetic tracer particles to determine RTD. The basis of this approach is related to Faraday's law of electromotive induction, which states that if a magnet moves towards or away from an electrical coil, an electric current is induced in the coil.

Another technique used by researchers to determine RTD of particles is fractional collection. In a study by Abdelrahim et al (1997), the experimental system consisted of a feed tank to store the working fluid, a particle tank connected to a three way valve, holding tube, and a screen to collect the particles.

Visual observation of colored tracer particles was applied by many researchers such as Fan and Fu (1996), Baptista et al (1996), and Sandeep and Zuritz (1994). Alhamdan and Sastry (1998) used videotaping in order to detect tracer particles flowing through a transparent SSHE and a holding tube. A mirror was used to monitor the particles that were blocked by the shaft of the SSHE. Residence times of the particles were obtained from the readings of the built-in digital stopwatch. Palazoglu and Sandeep (2002) used a digital imaging analysis technique. After videotaping of experiments, images captured from the entrance and exit of the holding tube were digitized by a media converter and downloaded on a computer. Residence time data were obtained by analyzing these images frame by frame.

Some recent studies by Sastry and Cornelius (2002), have revealed that use of magnetic resonance imaging or ultrasonic Doppler velocimetry have potential in determining RTD since they provide full velocity field information. However, more research is required to apply these methods to multiparticle food systems.

1.2.2.2 Research on RTD

Residence time distribution of liquids and particles depend on many system and process variables. The system variables are diameter, length, and geometry of the test equipment (holding tube or heat exchanger). Rheological properties, flow rate, temperature, and density of the carrier fluid, shape, size, density and concentration of the solid particles are the process variables (Ramaswamy et al 1995). A review of the effects of some of these parameters on RTD is presented below.

A study on RTD of water in a scraped surface heat exchanger (SSHE) was conducted by Abichandini and Sarma (1988). Dutta and Sastry (1990a, b) investigated velocity distributions of food particle suspensions in a straight holding tube. Both mathematical modeling and experimental studies were conducted to determine the average and fastest particle velocities.

Lee and Singh (1991) studied particle residence time distribution of particles in a horizontal scraped surface heat exchanger.

In another study by the same authors viz., Lee and Singh 1992, RTD of potato particles passing through vertical and horizontal scraped surface heat exchangers were mathematically modeled.

Ramaswamy et al (1992) developed an apparatus for particle-to-fluid relative velocity measurement. A transparent glass tube was used to monitor the flow of a particle. Experiments were conducted with water and 3 % starch solution at temperatures of 60, 80, and 90 °C. Potato, carrot, and polypropylene particles were used. It was found that the larger particles travelled faster than smaller particles due to a larger cross sectional area being available for high velocity streamlines. Starch solution was found to have a lifting effect on food particles, thereby yielding higher velocities. Velocities of food particles showed a greater variability than that of polypropylene particles. This was attributed to the smooth rolling of inert polypropylene spheres. When multiple particles of different diameters were used, the leapfrog phenomenon that was described by Dutta and Sastry (1990a, b) was observed. Smaller particles travelled as a string of particles, being pushed by the larger particles.

Abdelrahim et al (1993) used a fractional collection technique to collect residence time data of carrot cubes of 6 and 13 mm diameter in a starch solution. The consistency coefficient of the solution ranged from 0.052 to 0.657 Pa.s^n for a temperature range of 80 and 100°C. Larger particles were found to have higher residence times contrary to what Ramaswamy et al 1992) found. Ramaswamy et al (1992) found that larger sized particles traveled faster than their smaller counter parts. This discrepancy could be due to range of parameters such as flow rate or concentration of particles (5 % w/w versus single particle) used. As the temperature increased, the residence time of particles decreased due to increased turbulence as a result of the reduced fluid viscosity. The spread of residence times was characterized by the variance. It was found that variances increased with particle size and decreased with flow rate (15 and 20 kg/min), carrier fluid viscosity, temperature, and holding tube length (Ramaswamy et al 1992).

Sandeep and Zuritz (1994, 1995) studied the effects of several process parameters on residence times of multiple particles in a straight holding tube. Polystyrene spheres were used at 4, 7, and 10 % (v/v). Particles were close to neutrally buoyant with densities of 1,018 and 1,021 kg/m^3. The carrier fluid was CMC solution with consistency coefficient ranging from 0.6 to 1.4 Pa.s^n. Equations for dimensionless mean and minimum residence times were developed as functions of particle Reynolds number, particle concentration, and flow behavior index. Correlations for standard deviation of residence times involved particle size to tube diameter ratio. The results revealed that the standard deviation of residence times decreased with increasing flow rate, decreasing viscosity, increasing particle size, and increasing particle concentration. No channeling effects were observed which was opposite to what Dutta and Sastry (1990 a, b) found. This difference was attributed to very homogeneous suspensions due to the high particle concentrations used and the neutrally buoyant characteristics of the particles.

Effects of particle size and flow rate on residence time distribution in a curved section of holding tube were investigated by Salengke and Sastry (1995).

In another study, effects of particle concentration and the radius of curvature of the bend were analyzed by Salengke and Sastry 1996. Baptista et al (1996) investigated the effect of mixing particles with different characteristics viz., size and density on RTD.

Chandarana and Unverferth (1996) pointed out that if a thermal process was designed according to residence time estimates in the holding tube, excessive thermal treatment and loss of texture and nutritional qualities would result.

Fan and Fu (1996) studied the residence time of particles during vertical tubular flow. The radial migration effect caused by gravitational force was eliminated by using vertical holding tubes. Both single and multiple particle (up to 20%) experiments were conducted with water as the carrier fluid. Reynolds numbers varied between 6,300 and 26,000. Dimensionless mean residence times increased with Reynolds number. This was attributed to using particles that were heavier than the carrier fluid. Dimensionless standard deviations were found to decrease with increasing flow rate. It was observed that in vertical flow, particles moved at different speeds in the upward and downward parts of the tube. They reported that the standard deviation of residence times obtained in the vertical tube arrangement were lower (0.01 to 0.02) when compared to the results obtained from horizontal tubes (0.03 to 0.07) as studied by Sandeep and Zuritz (1995). However, more data would be required to verify the benefits of this vertical tube configuration.

Another study in a vertical pipe flow system was reported by Fairhurst et al (1999). Sandeep et al (1997) compared the residence time distribution of particles in conventional and helical holding tubes.

Another study by Sandeep et al (2000) was undertaken to model non-Newtonian two-phase flow in conventional and helical holding tubes. Palazoglu and Sandeep (2004) studied the RTD of particles in a single helical tube. Experimental parameters were flow rate and rheological properties of the carrier fluid, curvature ratio, particle concentration, and type of particle. Carrier fluid consistency coefficient which ranged from 0.05 to 0.20 $Pa.s^n$ did not have a significant effect on RTD. Mean and minimum residence times were found to increase as particle concentration increased from 4 to 12 %. Moreover, RTD was wider for higher particle concentration. This is in contradiction with what Sandeep et al (1997) concluded by increasing particle concentration from 3 to 10 %. Another previously reviewed study revealed no obvious effect of particle concentration within the range of 20 – 40 % (Salengke and Sastry 1996).

The effect of holding tube configuration on RTD was studied by Palazoglu and Sandeep (2002). Effect of curvature ratio, flow rate, carrier fluid consistency, and particle concentration were the other parameters studied. Polystyrene, acrylic, and a mixture of the two were used at concentrations of 4 and 12 % (v/v). A digital image analysis technique, which was developed by Simunovic (1998) was used to obtain residence time data. It was found that upon mixing, acrylic particles were lifted up and caught in secondary flow, which decreased the standard deviation of residence times. Increasing flow rate from 1.26 kg/s to 1.51 kg/s was more effective in reducing the standard deviation of residence times of acrylic particles due to the fact that neutrally buoyant polystyrene particles were already more uniformly distributed. RTD became narrower as particle concentration increased. This was contrary to the findings of Sandeep and Zuritz (1995), and Sandeep et al (1997), but in agreement with what Sandeep et al (2000) found.

The coiled flow inverter (CFI) is an innovative device, which has potential for the intensification of processes currently carried out in conventional mixers. Step response experiments were carried out in a CFI to study liquid-phase residence time distribution (RTD) for gas−liquid flow under the conditions of both negligible and significant molecular diffusion using Newtonian fluids. A total of 16 CFIs of different curvature ratios ranging from 6.7 to 20, dimensionless pitch from 1 to 2.5, and number of bends from 1 to 15 were investigated. The range of Dean Numbers for the gas and the liquid phase was varied from 235 to 1180 and 3.16 to 1075, respectively. The reduction in dispersion number is nearly 2.6 times for two-phase flow in CFI with 15 numbers of bends as compared to a straight helix under identical process conditions. A modified axial dispersion model is proposed to describe the RTD. The efficiency of the device is characterized by a mixing criterion as brought out by Subhashini Vashisth et.al (2009)**.**

The residence time distribution (RTD) in a new style tubular stirred reactor was studied. The fluid flow and RTD curves were solved using a commercial CFD code FLUENT 6.3(FLUENT Inc.).The results indicated that RTD was influenced by mass flow rate at inlet and stirring speed. The tubular reactor with a stirrer could improve the flow profile by narrowing the RTD curve, creating high Reynolds number, and avoiding back mixing (CAO Xiao-Chang et al 2008).

The model is built on the basis of the theory of Markov chains. The working space of a mixer is presented as a 2D array of perfectly mixed cells, through which the tracer travels according to the matrix of transition probabilities. The model covers the tendency of the tracer to upward/downward segregation. Finally, for a given flow pattern, the model allows calculating the RTD curves for various throughputs and for various properties of an imperfect tracer (Mizonov et al 2009).

1.3 OBJECTIVES OF THE RESEARCH

The objective of the study is improvement of prediction authenticity of theoretical methods by application of "up-to date" mathematical methods to simulate mixing process in static mixers of different operation principles. The aim is an application of the data obtained for development of methods of static mixer calculation (design). The objectives of the work in this thesis can be summarized as follows:

- To develop and validate a 2D model of turbulent flows in helical static mixers;
- To study the RTD of particles in the static mixer under different flow conditions.
- To demonstrate the benefits of helical static mixers from understanding the flow physics;

- To use the model to identify critical research issues as a guide to future research in this area.

The overall goal of this study is to develop a model that can predict efficiency of mixer operation in the conditions changed. Therefore, further theoretical and experimental research of mixing in static mixers is a representative scientific and technological problem. The Ricker model presented in this research is a part of a larger study on the use of static mixers for chemical reaction. It results in the objective of this study.

1.4 CONCLUSION

In this research, Residence time distribution (RTD) of particles is one of the critical factors. In a continuous flow system, some of the particles remain in the system longer than others, i.e. particles close to wall or in a dead space move much slower than those traveling along the centerline. Hence, the particles experience a distribution of residence times within the system. The Residence Time Distribution (RTD) is used to characterize the uniformity of the history of fluid elements in a static mixer. A similar history for all fluid elements in the flow is a desirable feature in order to provide the uniformity of the product quality. This can be achieved by a narrow distribution of the residence times for chemical reactors.

The tubular reactor provided with turbulence creators behaves as a plug flow reactor; since the elements perform the three types of mechanisms, namely flow division, reversal and combination even at low flow rates. Fluid flow through an annulus under turbulent condition also tends towards a plug flow situation. Hence one can expect in an identical way that the introduction of some turbulence promoters, in the annular space would also perform the same motions. No work seems to be reported in literature, concerning the effect of ‘turbulence creators’ inside an annulus. Hence with this aim, an annulus with ‘turbulence creators’ in the form of helical elements which are fitted on to the outer surface of inner tube, alternatively right and left handed in nature is constructed. Residence time distribution studies are performed. The above studies are also conducted in an identical annulus without mixing elements.

1.5 ORGANIZATION OF THE THESIS

In order to work on our objective, the following plan will be followed.

First, in Chapter 2, the basic principles of the static mixer are explained and an overview of the Mathematical model, theory and methods is given.

Reviews previous experimental and Mathematical studies of static mixers. The majority of the experimental studies of mixers are concentrated on design parameters for these mixers. These parameters may be immediately used in the design process. However, the existing experimental studies of static mixers provide very little information about how the mixers work and whether their performance is high or not. Unfortunately, the lack of detailed experimental data makes the validation of a Mathematical model of helical static mixers a challenging task.

In Chapter 3 determines the steady-state Residence Time Distribution, through stochastic modeling as a Markov process. The theory is applied to a real-life scenario on a steady state fluid flow in a tubular reactor.

Next, in Chapter 4 the experimental techniques are described and discussed.

The physics needed to study the flow in a mixer and describes the numerical method used to study the performance of a helical static mixer.

This chapter also presents a description of the Mathematical turbulence models, and also provides information about the stimulus response technique used in this study. Moreover, RTD modeling is discussed in this chapter.

Chapter 5 deals with a turbulent flow simulation of a static mixer and the static mixer performance predicted by Simulation is presented.

Chapter 6 presents a validation of the Mathematical modeling used in this research and presents results and discussion related to helical static mixer performance for Turbulent flow conditions and defines the scope for future research.