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Technical note

Radiotracer investigation in an industrial-scale oxidizer

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HIGHLIGHTS

- A radiotracer investigation was carried out in an industrial-scale oxidizer.
- The measured data were analyzed and flow parameters were obtained.
- The oxidizer behaved as an ideal stirred tank reactor.
- The results of the investigation were used to design of a new oxidizer.

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ABSTRACT

A radiotracer investigation was carried out in an industrial-scale oxidizer. The main objectives of the investigation were to measure residence time distribution (RTD) of organic process fluid, determine the mean residence time (MRT) and investigate the degree of axial mixing. Bromine-82 as p-dibromo biphenyl was used as a radiotracer for measuring RTD of the organic process fluid. The MRT of the fluid in the oxidizer was determined to be 390 min. An ideal stirred tank model with a plug flow reactor in recirculation stream was used to simulate the measured RTD data and was found suitable for describing flow in the system. Based on the model simulation the mean residence times in oxidizer and recycle stream were estimated. The results of the investigation showed that the oxidizer behaved as a well-mixed reactor whereas the recycle stream behaved as a plug flow reactor.

1. Introduction

A chemical industry in India operates an oxidizer for production of Dimethyl Terephthalate (DMT). The DMT plant was designed and setup by a foreign company and is about 25 years old. The schematic diagram of oxidation process and oxidizer for production of DMT is shown in Fig. 1. The DMT is used as raw material for manufacturing polyester fiber yarn, film, filament and engineering plastics. The DMT is produced by the well-known Witten Process that involves air oxidation of paraxylene (PX) and methyl para toluate (MPT) and esterification of the oxidate with methanol. The oxidation reaction takes place in the presence of catalyst. This reaction is an exothermic reaction and thus a large amount of heat is produced. This process is carried out in a vessel called an oxidizer that has an annual capacity to produce 165,000 t of DMT. The oxidizer is a bubble column reactor consisting of a vertical stainless steel cylindrical vessel of height of 27 m and diameter of 4.45 m. The operating volume of the reactor is 317 m^3 including volume of the vapor phase. During normal operation, about 20% of the volume of the reactor is occupied by the vapor phase (voidage) and remaining 80% is occupied by liquid (holdup). Therefore, the volume occupied by liquid phase in the reactor (V_o) was 253.6 m^3. The liquid reactants (*p*-xylene+methyl *p*-toluate+catalyst) are fed into the reactor at a feed rate of 40 m^3/h from the top section while the air is fed to the reactor from bottom at a flow rate of 28,500 N m^3/r through a sparger. The off-gas exits from top of the oxidizer. A fraction of the outlet flow i.e. residue is recycled back to the reactor after separation in a reactor. The flow rate of the recycled material was not known as there was no flow meter installed in the plant. The oxidation reaction is an exothermic reaction and significant amount of heat is generated during the reaction. In order to remove the excess heat, cooling water is continuously fed through the cooling tubes, provided in peripheral part of the oxidizer. The water is subsequently converted into steam. The reactor operates at a temperature of

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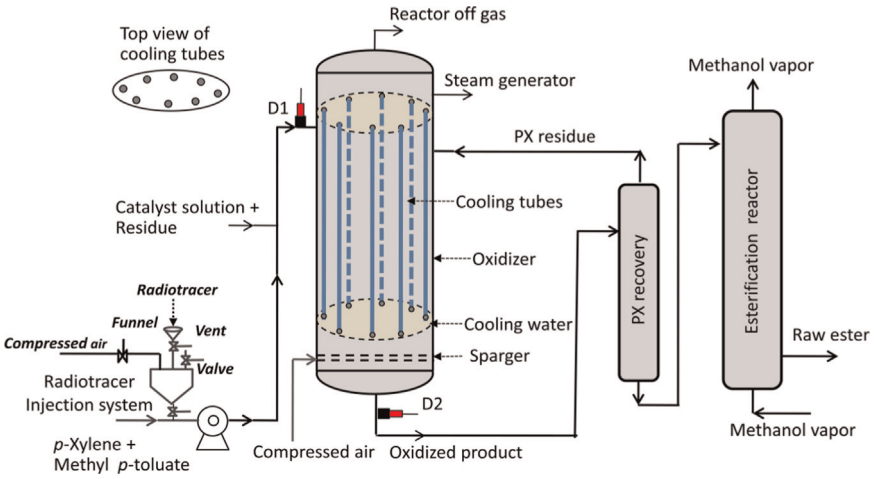

Fig. 1. Schematic diagram of the oxidizer and experimental setup.

165–170 °C and a pressure of 7 kg/cm^2.

The industry desired to enhance its production capability by indigenously designing and setting up another similar oxidation reactor. The oxidation process requires high degree of axial mixing of process fluids within the oxidizer with a specific value of MRT and the oxidizer is designed to behave as an ideal stirred tank reactor. Therefore, it was required to measure the mean residence time (MRT), degree of axial mixing of the process fluid within the existing oxidizer and identify any flow abnormality, if any. The obtained results were intended to be utilized for designing a new oxidizer. The concept of residence time distribution (RTD) is widely used to characterize flow in chemical reactors and measure mean residence time of process fluid (Danckwerts, 1953; Levenspiel, 1972). Various conventional tracer techniques are used for measurement of laboratory-scale reactors but the same cannot be used in industrial-scale reactors. However, radiotracer techniques are very effective tool to measure RTD of process material in pilot-scale as well as the full scale industrial systems because of their high detection sensitivity, "in-situ" detection, physicochemical compatibility, availability of wide range of suitable tracers, limited memory effect and utility in harsh industrial environment (Charlton, 1986; IAEA, 1990; Thyn et al., 2000; Pant et al., 2001, 2009a, 2009b; Pant and Yelgoankar, 2002). The paper describes the application of radiotracer technique in conjunction with a suitable mathematical model to estimate the hydrodynamic parameters of process fluid in an industrial oxidizer at normal operating conditions.

2. Radiotracer investigation

A radiotracer investigation was carried out to measure RTD, to determine MRT and to estimate the degree of axial mixing of the organic phase in the oxidizer. Two repeat tracer tests were carried out at identical process and operating conditions. Bromoine-82 as p-dibromo biphenyl was used as a radiotracer and about 1.5–2.6 GBq activity was used in the two repeat runs. The p-dibromo biphenyl salt was irradiated in DHRUVA research reactor located at Bhabha Atomic Research Centre, Trombay, Mumbai and dissolved in an organic solvent. The dissolved p-dibromo biphenyl was subsequently divided into two aliquots each of 5 ml volume and having activity about 2.6–2.8 GBq. A specially designed injection system as shown in Fig. 1 was connected to the PX feed line through a pressure tapping. The radiotracer was diluted in a volume of about 50 ml of PX and poured into the injection system. The radiotracer was instantaneously injected into the feed line using an air pressure of about 10 kg/cm^2 and monitored at two different locations in the oxidizer i.e. at the inlet feed line at the top end of the oxidizer and at the outlet of the oxidizer at bottom using two independent collimated NaI(Tl) scintillation detectors (1 in. × 1 in.). The detectors were connected to a computer controlled data acquisition system (DAS) set to record tracer concentration at an interval of one minute. Data was recorded till the radiotracer concentration reached to a natural background radiation level. The radiotracer could not be monitored on the recirculation line after PX recovery reactor due to some practical constraints. In addition to this, the possibility of conducting another independent radiotracer experiment to measure the recirculation flow rate (Q_r) was ruled out due to the non-availability of suitable locations for radiotracer injection and measurements in the plant.

3. Data analysis

The tracer concentration curves monitored at the inlet and outlet of the oxidizer were corrected for natural background, radioactive decay and tailing of the curve. The tail of the curves was distorted with fluctuations and was extrapolated with an exponential decaying curve (IAEA, 1996). The curve recorded at the inlet of the oxidizer (D1) was a sharp peak and was regarded as a Dirac's delta function. The time lag between the two curves monitored at the inlet and outlet of the oxidizer was about 1 min and can be considered negligible as compared to the mean residence time of the oxidizer. This implies that the radiotracer instantaneously mixes within the entire volume of the oxidizer and appears soon after injection of the radiotracer. Therefore, the normalized concentration curve monitored at the outlet of the reactor provides RTD of the oxidizer. The $C(t)$–t curve monitored at the outlet of the reactor was normalized by dividing each data point by the area of the curve and RTD curve, $E_{exp}(t)$ was obtained. Thus [1]

$$E_{exp}(t) = C(t) \Big/ \int_0^\infty C(t)\mathrm{d}t \tag{1}$$

The first moment of the RTD curve is the experimental MRT ($\bar{t}$) and is given as [1]

$$\bar{t} = tC\ \mathrm{d}t \Big/ \int_0^\infty C\ \mathrm{d}t \tag{2}$$

The experimentally determined MRTs are given in Table 1. The investigated oxidizer could be represented by a physiological