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Feasibility of Using Radioactive Particle Tracking as an Alternative Technique for Experimental Investigation in Bubble Column Reactor

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Abstract. Radioactive particle tracking (RPT) is one of the non-invasive techniques for monitoring and investigating multiphase flow system. This technique have been widely utilized in the field of chemical process engineering for better understanding and optimizing process hydrodynamics especially in the multiphase reactor such as bubble column reactor. Due to opaque nature of industrial process systems, especially in the case of multiphase flows, non-invasive methods based on ionizing radiation have been considered for evaluating the hydrodynamic parameters. The feasibility study of radioactive particle tracking techniques in quadrilateral bubble column reactor has been successfully achieved. The radioactive particle tracking facility and data acquisition system has been developed and experimental calibration using single particle radioactive particle ⁴⁶Sc to investigate dynamics behaviour of quadrilateral bubble column reactor is completed. The results indicated that there is back mixing behaviour in the bubble column process. The results also reported that the radioactive particle ⁴⁶Sc is still in good condition and there is no radiation contamination problem arises while performing radioactive particle tracking techniques. The RPT technique was performed to reveal the instantaneous velocity and time-averaged liquid velocity in the current bubble column reactor.

1. Introduction

Before the advent of the first Malaysian pool-type nuclear research reactor TRIGA MARK II at the Malaysia Nuclear Agency (formerly known as PUSPATI) in 1982, limited access to radioactive materials, limited source quantity and high-cost of radioactive materials have restricted Malaysian researchers in their efforts for establishing nuclear applications in oil and gas industries. Large-scale utilization of radioisotopes as research and industrial investigating tools began in the late 1990s when the Plant Assessment Technology Unit, Industrial Technology Division was established under Research and Development program of Malaysian Nuclear Agency. Since then, a radioisotope that emits γ -ray has been successfully used as nuclear gauges in research and industry. The versatility of nuclear gauges innovation offers better plant troubleshooting and problem-solving ability in oil and gas industry. Recent developments in the field of radioisotope application in industry have led to a renewed interest in studying process behaviors using radiotracer techniques. Industrial radiotracers emerged as powerful tools for troubleshooting and process optimization in multiphase flow systems. However, its performance is limited by the hydrodynamic parameters and its behaviors in complex multiphase reactors. Many critical and important information failed to be extracted using a conventional method. Thus, an alternative investigation technique is required to overcome this situation.



Due to opaque nature of industrial process systems, especially in the case of multiphase flows, non-invasive methods based on ionizing radiation have been considered for evaluating the hydrodynamic parameters of multiphase reactors. These parameters include flow regimes, phase holdup, phase distributions, velocity profiles, mixing patterns and turbulence fluctuations. Thus, the demand for reliable experimental techniques to provide non-invasive, spatially, and temporally resolved data of single-phase and multiphase systems is required to assess the accuracy of complex models. The role of industrial radiotracer technology in industries has received increased attention across some disciplines in recent years. In the last decades, a significant effort to utilize γ -ray emission techniques for measuring positions and velocities in three dimensions in multiphase system reactors has been highlighted. One of the non-invasive techniques for monitoring multiphase flow system is known as a radioactive particle tracking (RPT) technique. This technique has been implemented extensively to investigate hydrodynamics characteristic in a great variety of multiphase flow systems [1].

Radioactive particle tracking (RPT) techniques has been widely utilized in the field of chemical process engineering for getting better understanding and optimizing process hydrodynamics especially in the multiphase reactor non-invasively. This technique applied a simple approach where an array of scintillation detectors is used to detect emitted isotropic γ -rays signal from radioactive particles over a sufficiently long period of time as to track the motion of that particle at a specific position and location. The radioactive particle position at certain time can be reconstructed from the widely used rigorous phenomenological model which correlates the number of photons detected and effectively measured by a scintillation detector and the positions of the emitting γ -ray radioactive particle. This strategy also involves the reconstruction of the tracer location histories known as calibration map from its real position coordinates by solving a minimization problem between the experimental and calibrated data. This is done based on tracking the motion of a neutrally buoyant single radioactive particle in order to visualize and validate the flow inside a specific volume [1,2,3,4].

There are relatively few historical studies in this technique with the first detailed study of radioactive particle tracking conducted by Kondukov *et al.* [5]. However, there is an inadequacy in his data acquisition system where quantitative results were failed to be secured. In his work, Kondukov *et al.* [5] traced a radioactive particle inside gas-solid phases in fluidized bed reactor by using 6 units scintillation detectors. Similar numbers of detectors were also used by Meek [6] for tracing solid material in turbulent liquids using different experimental design with detectors mounted on an axially moving carriage to move along with the solid particle. Unfortunately, the carriage failed to maintain its speed with tracer particle leading to data losses. Another researcher, Lin *et al.* [2] introduced first innovative computer-aided radioactive particle tracking known as CARPT with low-resolution detection from 12 unit of detectors analogue processing to investigate the solid dynamics inside the gas-solid phase of fluidized bed reactor. From then on, system transformation to the advanced radioactive particle tracking system becomes the center of attention for better hardware improvement such as those related to the data acquisition systems. The data acquisition system developed by Lin *et al.* [2] has been upgraded by Moslemian [7] where the digital pulse counts meter were introduced and has been successful in providing high-speed counting rates.

In 1991, Devanathan [3] utilized the radioactive particle tracking for investigating the first liquid phase dynamic in a gas-liquid column reactor test rig by introducing a neutrally buoyant radioactive particle [3,8]. Subsequently, the same principles have been applied by Yang, [9] to perform better numerical calculation using a weighted least-squares method in particle tracking post-processing techniques, extensively. The position rendering method proposed by Yang, [9] promoted another invention on the rigorous phenomenological relation generated using Monte Carlo simulation by Larachi *et al.* [10] followed by the development of enhanced algorithm with direct back-propagation neural network model [11].

Much of the literature since the mid-1990s emphasizes the use of non-invasive radioactive particle tracking techniques for the characterization of multiphase reactors. Research into radioactive particle tracing applications in multiphase flow system has a long history. There are a large number of published studies that describe the uses of radioactive particle tracking technique for gas–solid–liquid phase

investigations in fluidized bed reactor [1,12,13,14,15,16,17,18,19,20,21,22,23,24], solid phase in cylindrical tumbler reactor [25,26], gas–solid and gas–liquid phase in risers [27,28,29,30,31], liquid phase in circulating draft tube photobioreactor [32], solid–solid phase in V-blender reactor [33], and pebble bed reactors [34,35]. However, there is very little published research on the gas-liquid phase in quadrilateral bubble column reactors which need to be comprehensively investigated and examined using radioactive particle tracking technique.

Surprisingly, there have been no preliminary works reported for the application of radioactive particle tracking techniques in Malaysia. The primary challenge faced by researchers in Malaysian is the limited capability to produce or purchase radioactive material at a reasonable cost. In addition, the radioactive particle tracking hardware and portable data acquisition system is currently not suitable for field testing in the petrochemical industry. This scenario indicates the need to set up innovative RPT facility for hydrodynamics investigation of industrial-scale reactors in Malaysia. Therefore, this study seeks to remedy those situations by designing and utilizing radioactive particle tracking as an alternative technique for the first experimental investigation in bubble column reactor in Malaysia. Furthermore, this study also set out to gain further understanding of the feasibility of using this alternative method for gas-liquid phase behavior. A specialized radioactive particle which neutrally buoyant in water phase will be used together with the fixed radiation detectors around the quadrilateral bubble column reactor. This radioactive particle tracking technique is using the simulated calibration map using MCNPX code to track the particle position in order to reconstruct the particle trajectory and visualize the velocimetry behavior of the water phase in bubble column reactor. The development of data acquisition system hardware and software will also be discussed.

2. Methodology

2.1. Experimental Setup

The experiments were carried out in a quadrilateral bubble column reactor test rig with an internal diameter of 200 mm and a height of 1800 mm. Figure 1 shows a schematic diagram of a quadrilateral bubble column reactor used for radioactive particle tracking measurement. The quadrilateral column was fabricated using Perspex® acrylic glass (density = 1.18 g/cm³) with inner cross-section equal to 200 mm x 200 mm and wall thickness equal to 3 mm. The column is filled up with 0.04 m³ normal tap water of ambient pressure and at a room temperature. The height of the fluid volume (water level) referred to the stationary liquid level was fixed at 1000 mm. The air as a gas phase was supplied from centralized compressors with a working pressure of 1.5 MPa. A pressure regulator and flow meters regulated the air flow rate at a fixed rate of 20 liters per minutes. Air is purged into the reactor to create air bubbles for aeration from the bottom by passing through perforated air distributor known as sparger plate at the bottom of the bubble column reactor. The sparger design D used in this study has a porosity of 0.18% and the total opening cross-sectional area of 70.86 mm². It consists of 361 holes of 0.5 mm diameter with the triangular pitch of 1 cm apart.

Radioactive particle tracking facility consists of an array of radiation detectors and data acquisition systems. Ten (10) units 2 inch x 2 inch (5.08 cm diameter x 5.08 cm long) crystal of sodium iodide activated with thallium, NaI (Tl) scintillation detectors (crystal density = 3.67 g/cm³) are strategically fixed around the bubble column reactor as can be seen in Figure 1. The detectors are positioned on five planes alternately, and the distance between the consecutive planes is equal to 20 cm. The detectors are held by aluminium extrusion profile bar structures which in turn are supported on stainless steel round bars with dimension 3.81 cm x 3.81 cm positioned at 90° intervals around the column. Two detectors are placed at one level to face each other, surrounding the column with 180° spacing. The distance between the surface of the detector and the column wall is fixed at 5 cm equally. The arrangement of the detectors in each plane is shown in Figure 2.

When radiation passed each scintillation detector, the radiation counts were collected instantaneously and recorded into a computer. The hardware components used for this experiment is a multi-channel

analyzer Ludlum Model 4612 Counter, and a 12-detector single channel analyzer (SCA) with RS232 control to compute the signals from the scintillation detectors. Block diagram of the data acquisition system is shown in Figure 3. The desktop computer or industrial durable notebook is used to control all the parameters, obtain and display data, and extract results. The physical picture of radioactive particle tracking setup for bubble column reactor can be seen in Figure 4. This versatile data acquisition hardware system set up is safe to be used in oil and gas industries. The compact set of the multi-channel analyzer, scintillation detector and durable notebook are using minimal electrical power source. Therefore, the set up can reduce the potential to induce spark and ignition that increase the risk of fire and explosion inside refinery plants.

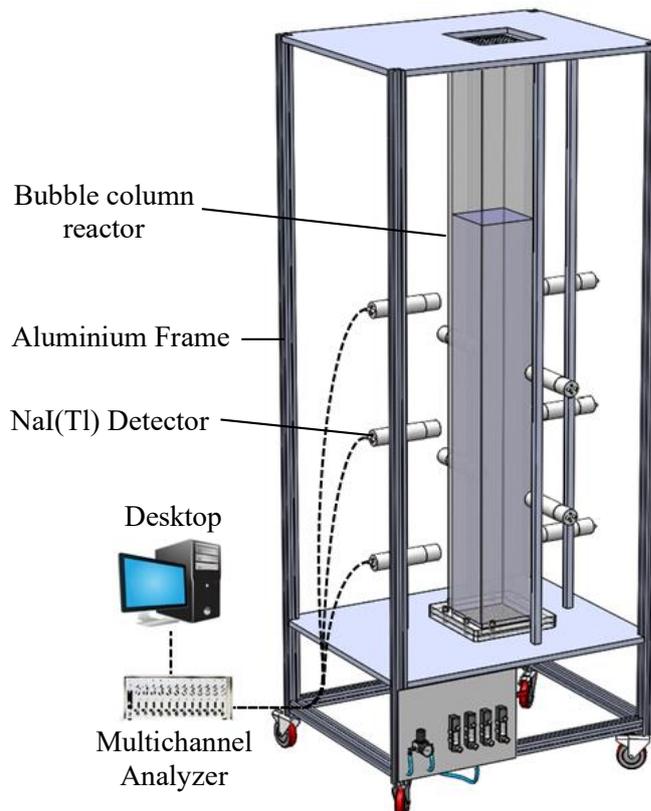


Figure 1. Schematic diagram for the quadrilateral bubble column reactor and radioactive particle tracking setup.

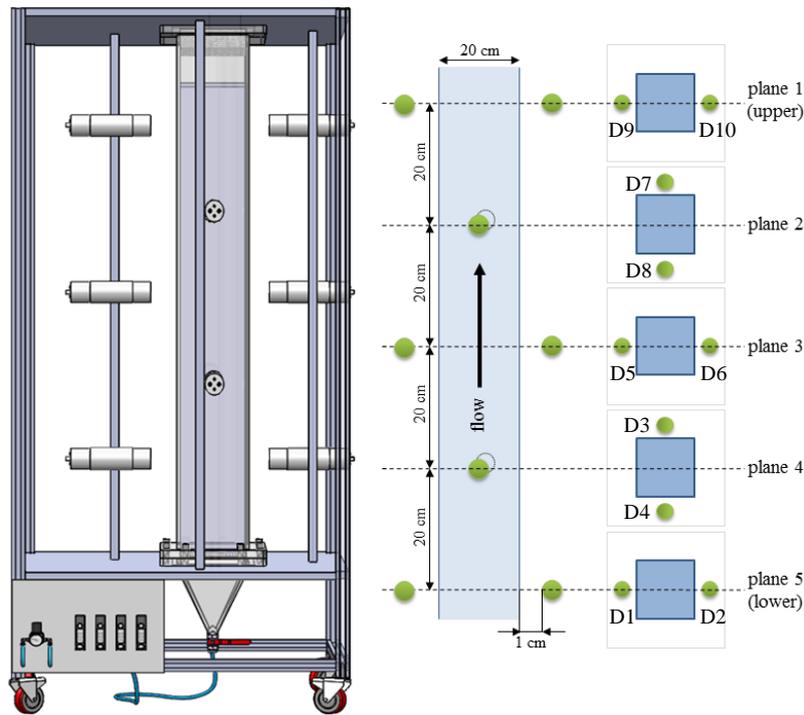


Figure 2. The arrangement of the scintillation detectors in experimental setup

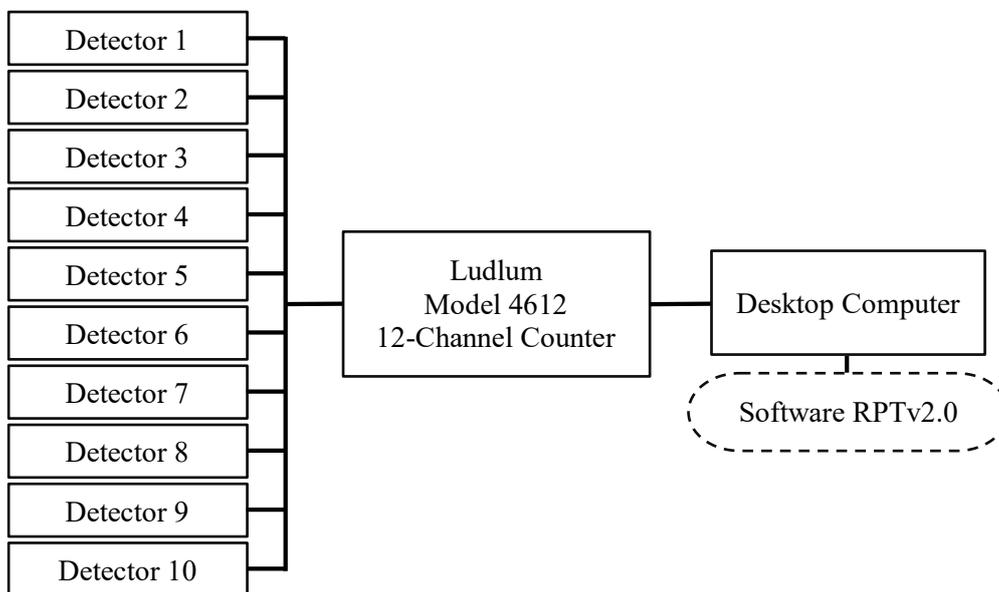


Figure 3. Block diagram of the data acquisition system.

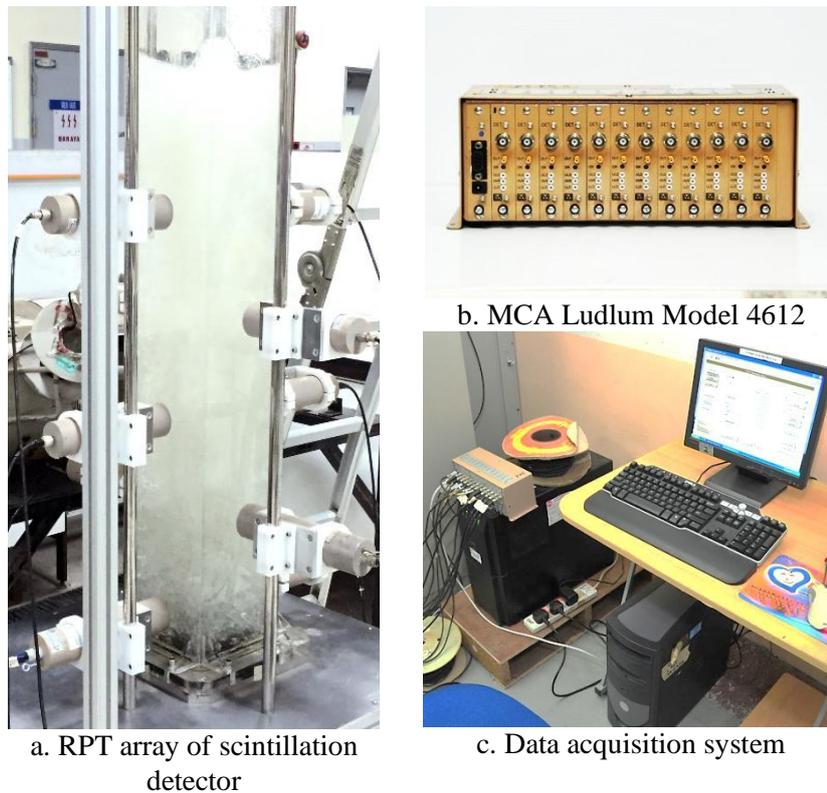


Figure 4. Physical pictures of RPT experimental setup.

2.2. The Radioactive Particle

Due to various advantages, a scandium radioactive particle with an activity of 4 mCi was selected for this experiment. This selection is made based on the fact that the radioisotope ^{46}Sc has nuclear transitions emitting gamma radiation with sufficient energy for a radioactive particle tracking application. Scandium has a half-life of 83.8 days and presents two photopeaks, one at 889 keV and one at 1121 keV. In radioactive particle tracking technique, a single tiny radioactive particle that is naturally buoyant is used for tracking the motion of liquid phase in bubble column reactor. Enough space is required to maintain the buoyancy of the particle equivalent to water density. Taking this fact into consideration, a hole is made in polyethylene bead that contains the scandium glass and the empty gap of the hole is filled sufficiently with the Araldite epoxy glue to prevent radioisotope dilution, dispersion, and leakage that may lead to radioactive contamination inside and outside the reactor. The air gap and amount of the glue inside the particle are adjusted to match the density of water. About 20 mg scandium glass (Sc_2O_3) was encapsulated in polypropylene beads sphere particles of diameter 2–3 mm so that the effective density of the tracer particle is closely matched with the density of liquid phase. Final target density practically close to the water density of 1.001 kg/m^3 at ambient temperature where the neutrally buoyant particle is achieved. The γ -ray emitter ^{46}Sc radioactive particles are activated by thermal neutron activation in a nuclear reactor. In this study, the particle was irradiated at 750 kW for 6 hours with neutron flux density of 1.2×10^{12} thermal neutrons per cm^2/s inside the core of TRIGA-Mark II reactor at the Malaysian Nuclear Agency.

2.3. RPT Data Acquisition Software

Detection of single radioactive which emits gamma radiation was achieved by the use of a scintillation detector without collimator positioned around the bubble column. The response signals produced by the detector were recorded continuously at the specific interval period of time. Data acquisition companion software for acquiring data and recording response signals from sets of scintillation detector in

radioactive particle tracking experiment initially developed by Rahman *et al.* [36] for study the hydrodynamic behaviour in quadrilateral bubble column reactor. National Instruments LabView 8.6 system design software has been utilized to develop radioactive particle tracking data acquisition system software. This system software consists of an interactive scanning control graphical user interface (GUI), a data acquisition from 12-channel RS-232 serial interface counter, and a multichannel analyser (MCA). Rahman *et al.* [36] reported that the second version of the software was successfully upgraded and currently known as RPTv2.0. The graphical user interface (GUI) for this software includes three operating modes that consist of system setup and configuration mode, scanning mode and multichannel analyser, upper-level discriminator, and lower-level discriminator (MCA & LLD/ULD) setup mode.

The system setup and configuration mode enabled the configuration of COMM port for communication between the multichannel analyzer and the desktop host communication port. This can be seen in Figure 5. Operating system input setup such as total scanning time, the configuration setting, and output files setup are available in this mode. Before running the data acquisition mode or scan mode, a suitable window of the region of interest for gamma radiation spectrum must be set in the software. Thus, the MCA and LLD/ULD setup mode shown in Figure 6 can be used to configure the lower and upper levels of gamma spectrum with communication between the software and a multichannel analyzer. This mode is designed to help the user in selecting the appropriate values for LLD and ULD parameters between 0 to 3300 millivolts. The graphical user interface of LLD/ULD adjustment of gamma spectrum result for ^{46}Sc radioactive particle tracer is shown in Figure 7. The graphical user interface for scanning mode is shown in Figure 5. In this mode, the integration time (counts rate) and the total period of acquisition time both have to be determined before starting the data acquisition process. The integration time describes the duration of each data count and period of acquisition means the total time of the data acquisition process. Ludlum Model 4612 multichannel analyser has the capability to obtain the data counts at -higher speeds up to 50 ms per count. The scan mode also allows user to control the start and stop the scanning process. The updated count signals are displayed with respect to the integration time and the scanning process is continued until desired counting time is reached. Immediately upon the completion of data acquisition process, a pop-up graphical user interface will inform the user to save the acquired results in the specified folder location [36]. Figure 8 shows the flowchart for data acquisition system process by utilizing RPT hardware and software.

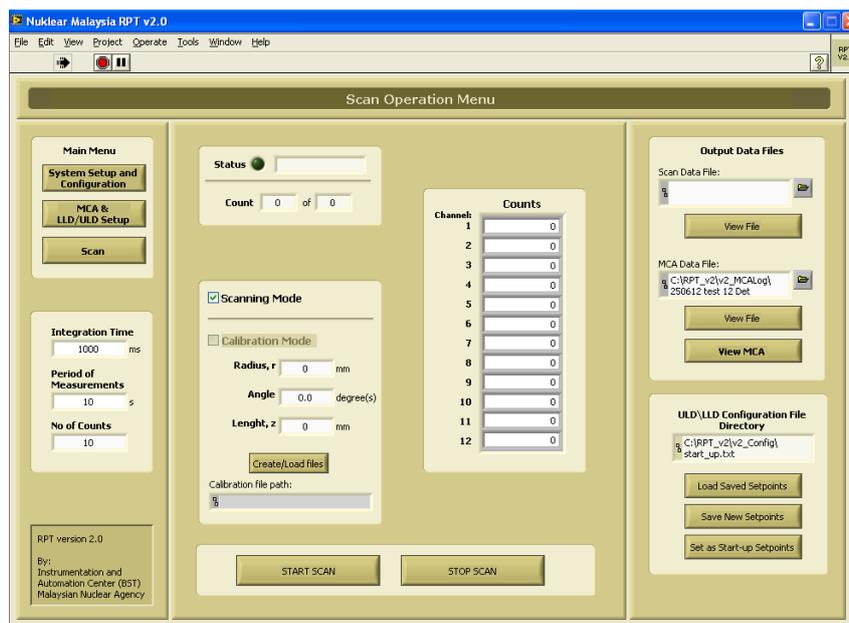


Figure 5. The graphical user interface (GUI) for RPT software version 2.0.

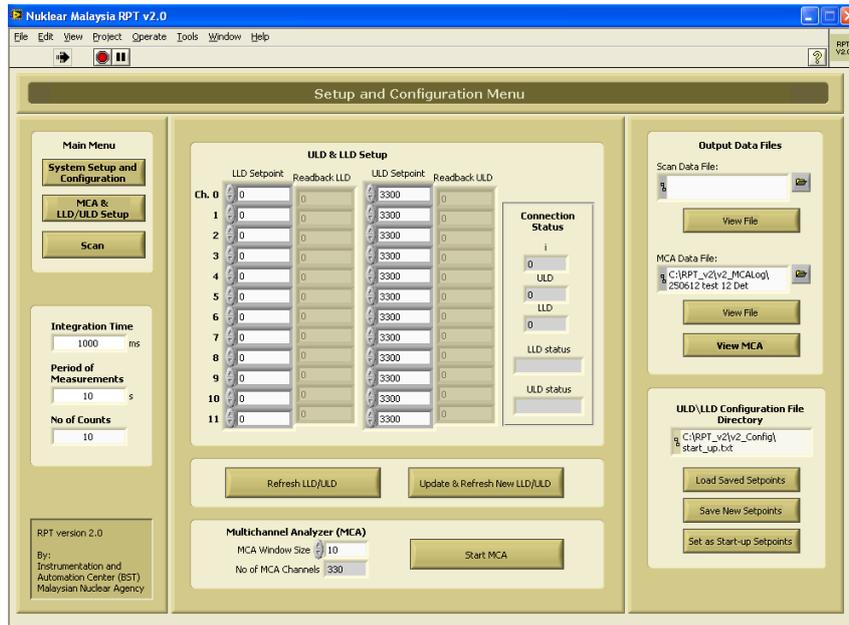


Figure 6. The MCA and LLD/ULD setup mode GUI.

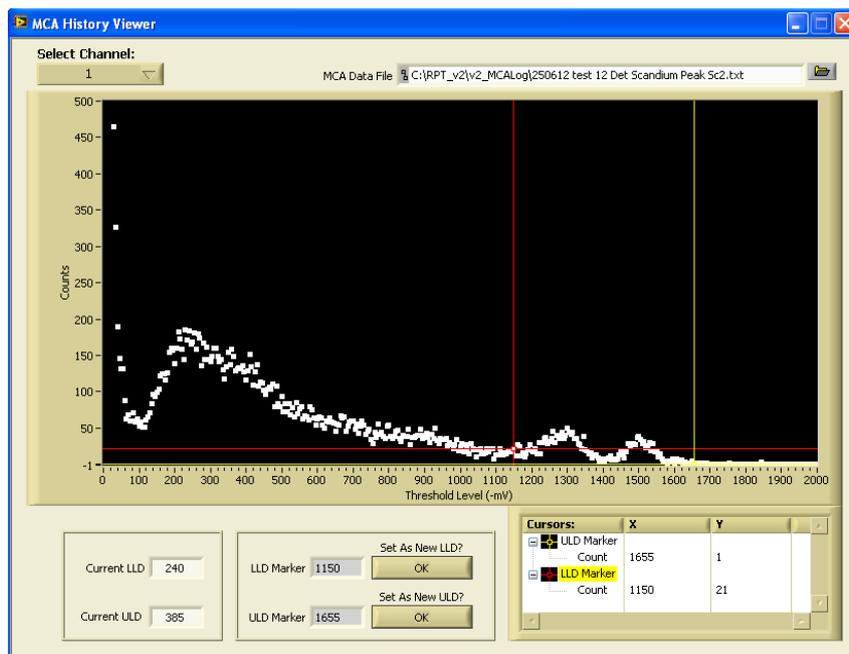


Figure 7. LLD/ULD marker adjustment in gamma spectrum results for ⁴⁶Sc.

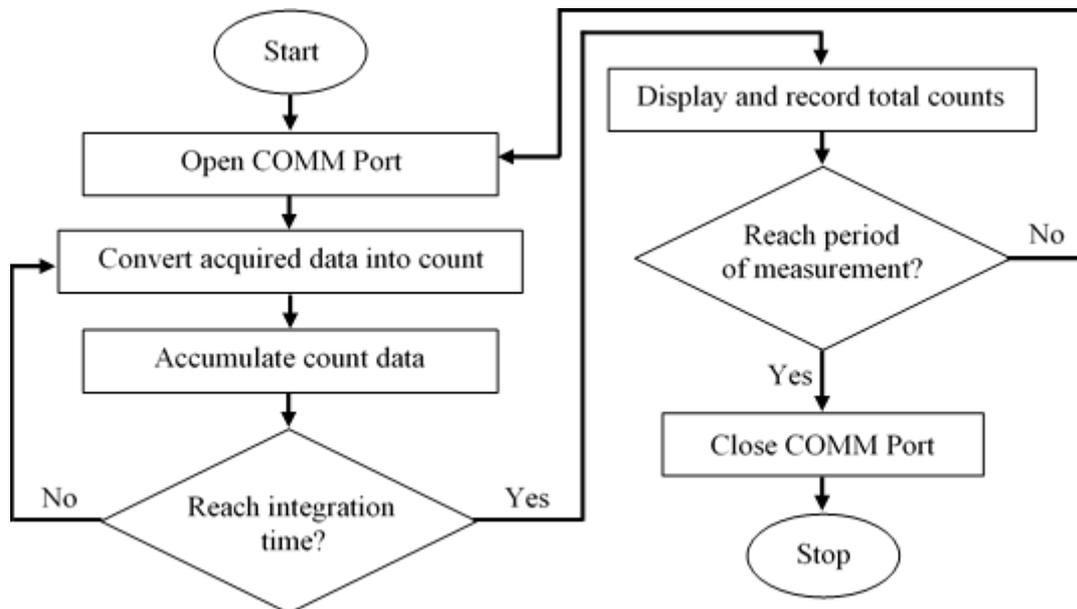


Figure 8. Flow chart for data acquisition system process.

3. Results and Discussions

The present study was designed to determine the feasibility of using radioactive particle tracking as an alternative technique for hydrodynamics behavior investigation in bubble column reactor. The radioactive particle tracking facility has been successfully developed. The facility includes the system hardware that consists of pilot-scale quadrilateral bubble column reactor, ten sets of scintillation detector with special detector holder, and data acquisition system that used compact and versatile multichannel analyzer. One of the primary concern in using alternative radiotracer method is the radiological safety to radiation workers and members of public. In this study, encapsulated ^{46}Sc radioactive particle inside polypropylene bead was successfully prepared to prevent contamination inside the reactor vessel.

Before the RPT experiment was carried out, the position of the particle has to be reconstructed accurately from the analyzed radiation signals. The particles will be placed at known positions depending on the size and volume of the vessels to be mapped. There are few methods used to calibrate the particle position in radioactive particle tracking technique. First method uses a calibration device which was able to calibrate the locations of radioactive particle in angular, axial and radial directions using the automatic computerized device. This device is known as computer-aided radioactive particle tracking (CARPT) [13]. The second method is using position reconstruction by utilizing the Monte Carlo simulation code for solving a minimization problem between measured counts and a rigorous phenomenal model that relates the position of the emitter to the number of counts recorded by each detector surrounding the system. The detectors measure different levels of radiation depending on the position of the emitter. Using an appropriate model, the exact location of the tracer can then be obtained.. Generally, in Monte-Carlo based reconstruction, the intensities of γ -rays emitted by tracer particle at specified locations are documented by positioning scintillation detectors around the vessel during the calibration stage.

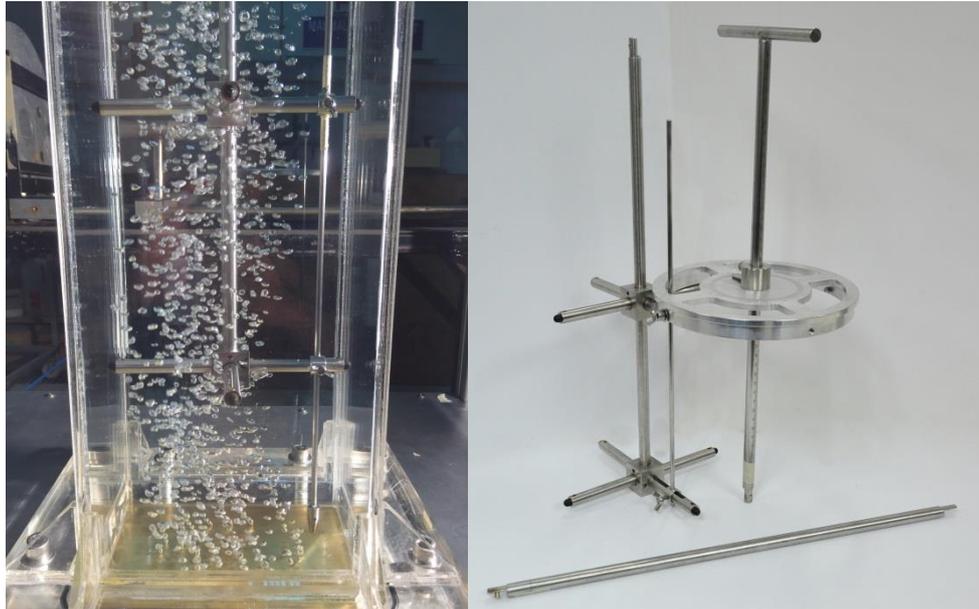


Figure 9. Manual radioactive particle calibration holder devices.

In this study, manual calibration process using special equipment is chosen because automatic calibrations devices are currently not available for radioactive particle tracking. In order to track particle inside the bubble column reactor accurately, a conventional benchmark experiment was carried out using manual calibration equipment to validate the particle position reconstructed by Monte Carlo simulation. This equipment was fabricated using stainless steel with single radioactive particle holder as can be seen in Figure 9. The calibration equipment was designed to reconstruct particle histories. Figure 10 indicates that distance versus radiation signals relationships known as calibration curve for the detector provides the basis to reconstruct the tracer particle instantaneous position. A comparison between the simulated particle positions reconstructed using Monte Carlo algorithm and the particle positions obtained experimentally is shown in Figure 10. The comparison shows a great matching between these two sets of coordinates. MCNPX code was applied to calculate a map of counts containing a total of 2600 grid points as a function of particle coordinates. In total, 26000 data sets have been successfully obtained with ten detectors were used for the reconstruction of instantaneous positions with the aid of the 2600 known coordinates.

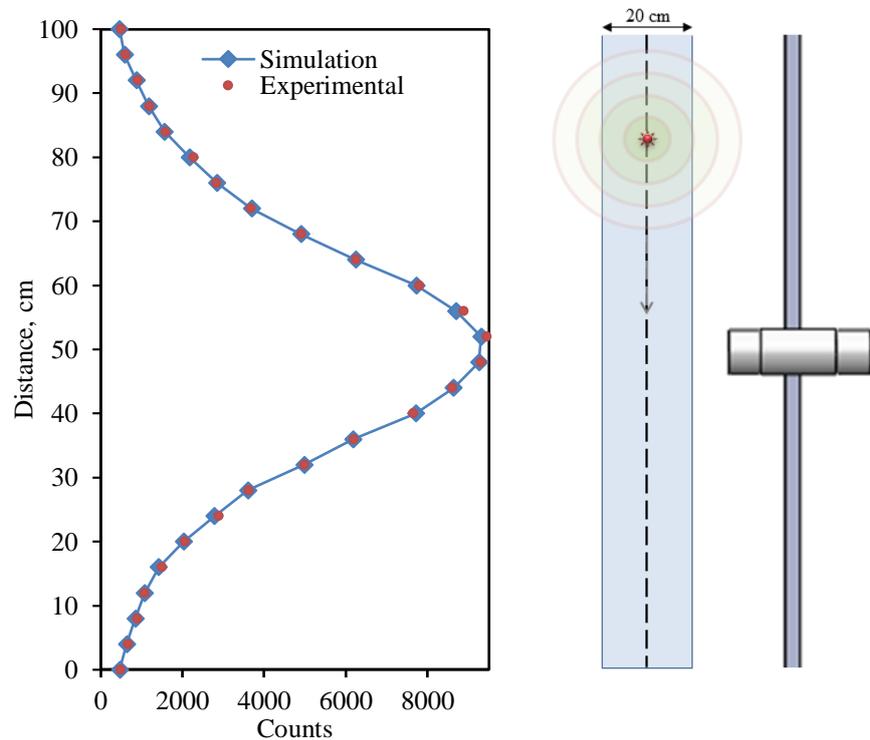


Figure 10. A comparison between MNCPIX simulation and experimental distance counts rate measurement for Detector 5.

Upon completing the calibration step, the neutrally buoyant radioactive tracer particle will be left freely moving with the liquid phase inside bubble column reactor while the scintillation detectors collect count intensities at a suitable frequency (10-20 Hz) depending on the flow environments and the activity of the radioactive particle. A sampling frequency of 20 Hz and total acquisition time of 2.5 hours is used in this study. This is because the homogeneous regime conditions in the bubble column reactor with superficial gas velocity is 0.0083 m/s. Figure 11 shows the data acquisition results for ten detectors at different planes and radioactive particle ^{46}Sc axial trajectories for the acquisition time 2.5 minutes was illustrated at different levels. By using the calibration curve and the reconstruction algorithms, the Lagrangian trajectory of the radioactive tracer can be reconstructed and the result is as shown in Figure 12 (a) and (b). The experiments will be operated for a sufficient time to allow the radioactive particles numerously explore the whole region of bubble column reactor so the time-averaged velocity in each compartment reaches a plateau. This will ensure enough statistics related to radioactive tracer particle trajectories, which represents the liquid dynamics of the bubble column reactor.

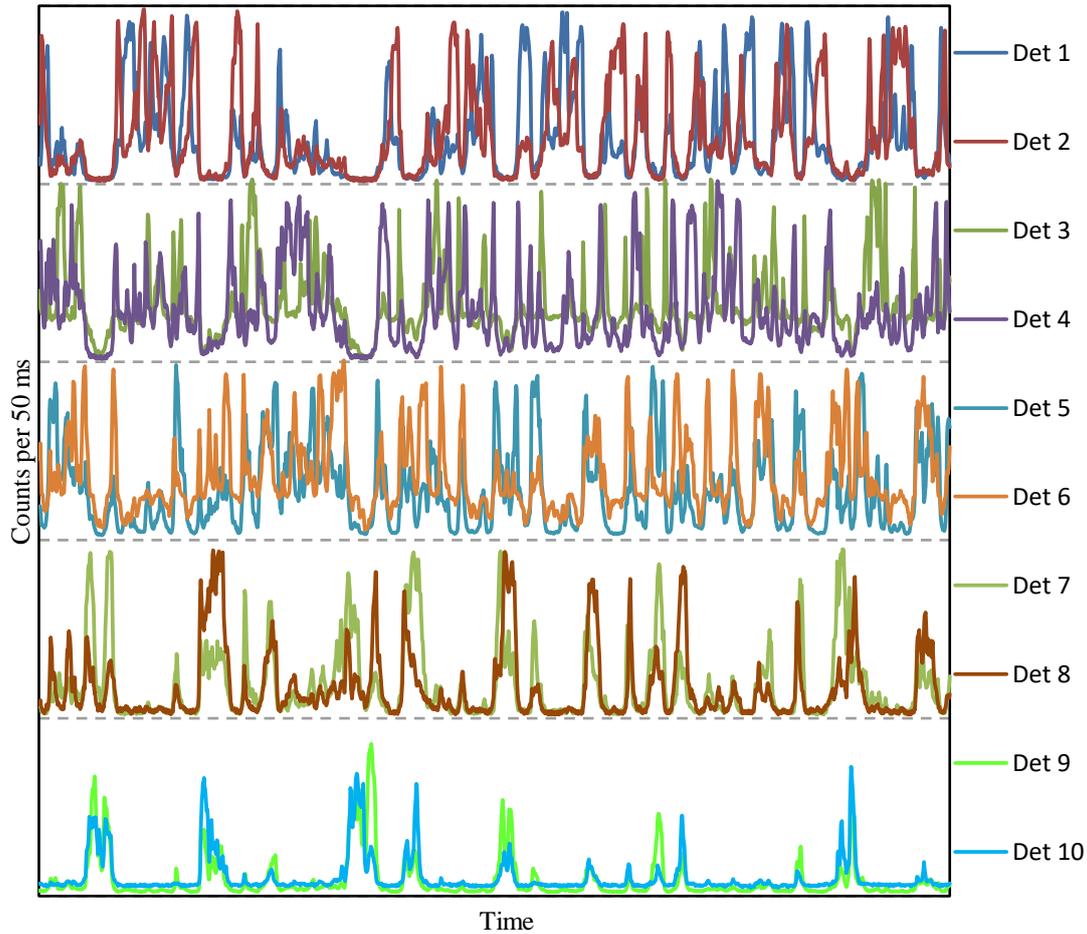


Figure 11. Radioactive particle ^{46}Sc axial trajectories for the 2.5 minutes at different detector level.

Once the distance of the particles from the set of detectors is evaluated, a weighted regression scheme is used to estimate the position of the particle at a given sampling instant in time. Thereby a set of instantaneous position data is obtained that gives the positions of the particle at successive sampling instants. Time differentiation of the successive particle positions yields the instantaneous Lagrangian velocities of the particle as a function of time and position of the particle. From the Lagrangian particle velocities, ensemble averaging is performed to calculate the average velocities of the liquid. For Cartesian coordinate system, the instantaneous velocity (u_i) and mean velocity (u) were calculated by determining the magnitude of the velocity vector (speed) as follow:

$$u_x = (x_2 - x_1)/(t_2 - t_1) \tag{1}$$

$$u_y = (y_2 - y_1)/(t_2 - t_1) \tag{2}$$

$$u_z = (z_2 - z_1)/(t_2 - t_1) \tag{3}$$

$$u_i = \left\{ \frac{(x_2 - x_1)}{(t_2 - t_1)} \right\} i + \left\{ \frac{(y_2 - y_1)}{(t_2 - t_1)} \right\} j + \left\{ \frac{(z_2 - z_1)}{(t_2 - t_1)} \right\} k \tag{4}$$

$$u = \sqrt{u_x^2 + u_y^2 + u_z^2} \tag{5}$$

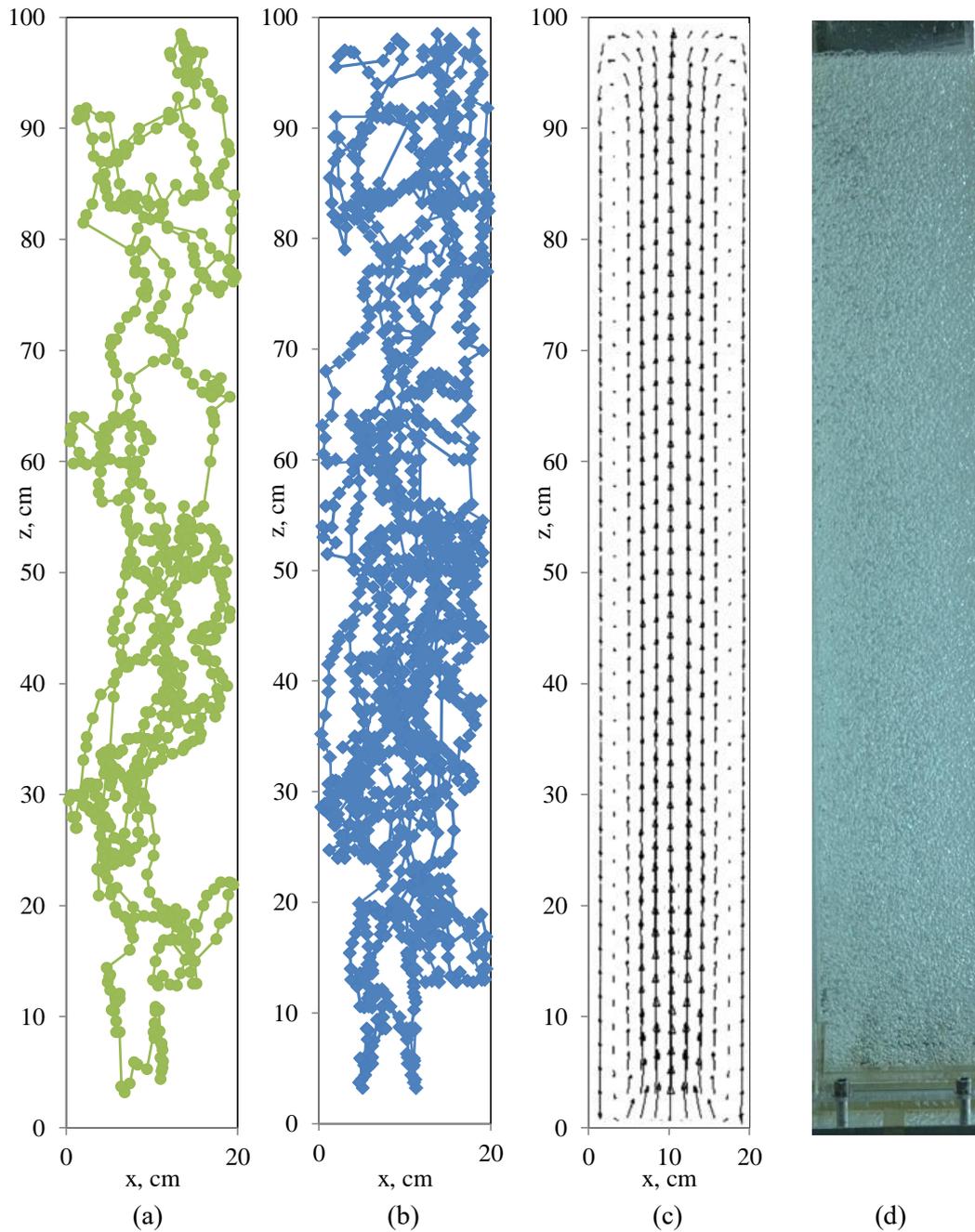


Figure 12. Lagrangian trajectory of radioactive particle ^{46}Sc in bubble column reactor with superficial gas velocity 0.083 m/s for (a) 25 sec acquisition time (b) 75 sec acquisition time. (c) Time-averaged velocity vector plots. (d) A graphical image of the dispersed bubble for sparger design type D.

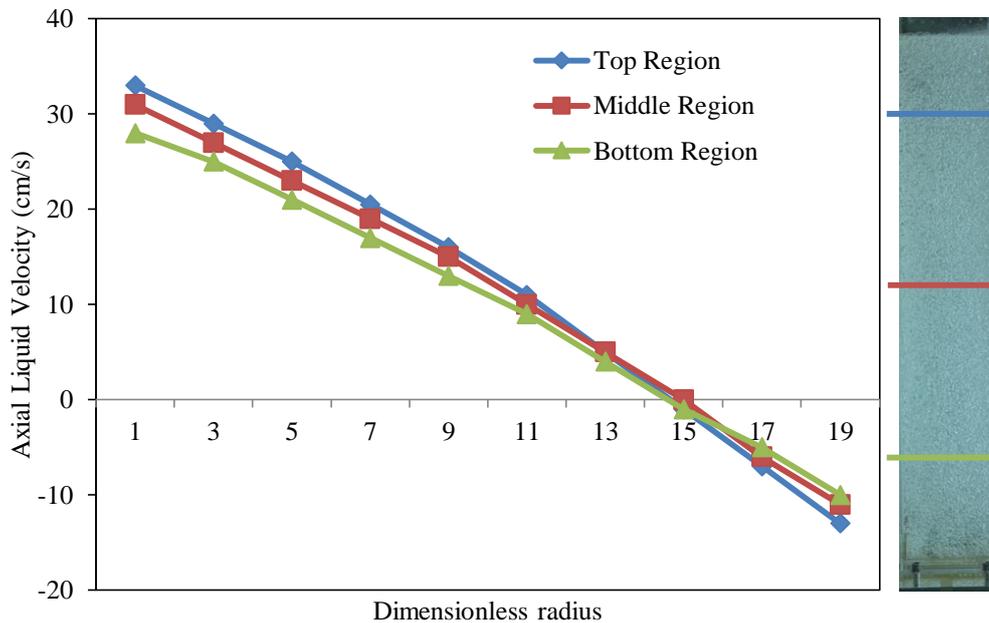


Figure 13. Comparison of axial liquid velocity at different region of column.

The instantaneous liquid velocity in Figure 12(c) has been calculated by time differentiating between two successive locations of the tracer particle. The time-averaged liquid velocity is obtained by averaging the instantaneous velocity of each cell by taking the mean of the number of occurrences obtained for each sampling compartment as shown in Figure 13. The results presented the comparison between bubble column region at top, middle and bottom of the vessel. Thus, this study concluded that the alternative RPT technique is also capable of providing information about the fluid dynamic parameters.

4. Conclusions

The feasibility study of radioactive particle tracking techniques has been successfully completed. The radioactive particle tracking facility has been developed and experimental calibration using single particle radioactive particle ^{46}Sc to investigate dynamics behavior of quadrilateral bubble column reactor has been accomplished. The RPT technique was performed to reveal the instantaneous velocity and time-averaged liquid velocity of bubble column reactor. The results indicated that there is back mixing behavior in the bubble column process. The results also reported that the radioactive particle ^{46}Sc is still intact and there is no radiation contamination problem arises while performing radioactive particle tracking techniques.

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