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Industrial Radiotracer Technology for Process Optimizations in Chemical Industries – A Review

Mohd Amirul Syafiq MOHD YUNOS[a,b*], Siti Aslina HUSSAIN[b],
Hamdan MOHAMED YUSOFF[b], Jaafar ABDULLAH[a]

[a]Malaysian Nuclear Agency, Bangi, 43000 Kajang, Selangor, Malaysia
[b]Faculty of Engineering, Universiti Putra Malaysia, 43400 UPM Serdang, Selangor, Malaysia
syafiq@nuclearmalaysia.gov.my*, aslina@upm.edu.my, hamdan@upm.edu.my,
jaafar@nuclearmalaysia.gov.my

Abstract - Radioisotope techniques are constantly and extensively used all over the world as a method to identify process systems malfunctions in various industries without requiring the shut down of the processing plant thus leading to high economical benefits to the plant owner. Different aspects of industrial radiotracer technology for troubleshooting, process control and optimization are evaluated through an exhaustive literature survey. The review covers the advantages of radiotracers, most commonly used radiotracers in industry for specific studies, applications of radiotracer techniques in various chemical industries, the design of radiotracer technology experiments, radiation detection and data acquisition in radiotracer technology as well as radiological safety aspects. Two industrial radiotracer techniques of residence time distribution (RTD) measurements and radioactive particle tracking (RPT) are discussed. The design of radiotracer technology experiments are also divided into two categories - radioactive particle tracking applications and residence time distribution applications.

Keyword: Industrial radiotracer, Industrial process, Process optimization, Residence time distribution, Radioactive particle tracking.

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Introduction

Optimization has been defined as the selection of the best method to achieve an objective. In the chemical industry, the ultimate objective is monetary gain. So optimization may be said to be the process of matching the production to the needs of the marketplace at the lowest unit production cost. Thus, the successful operation of a plant depends on the early identification of the problems for remedial action to be conducted and further optimization of the plant performance can be established (Pant et al., 2001).

This process may be extremely complex and it is much an art as science is involved, as well as factors such as economic judgement and commercial acumen. It also involves technical and scientific judgements such as selection of the most appropriate process technology, choice of optimal unit design and control of plant operations to achieve maximum benefit. This latter category of activities, the technical aspects of the general optimization problem, encompasses what is usually referred to as process optimization and it is in this sphere of activity that radioisotope technology can make important contributions.

Moreover, radioisotope techniques are constantly and extensively used all over the world in the area of identifying malfunctions in process systems in various industries without the need to shut down the processing plant, thus leading to high economic benefits to the plant owner. The phrase 'radioisotope technology' has been used deliberately rather than radiotracer technology. Radioisotope techniques based on sealed sources of radiation are often neglected in the literature of this subject even though the said application shows great effect in complementing radiotracer studies or as investigative tools in their own right. Radioisotope technology can assist all stages of process development and operation. Thus, radiolabeled compounds are widely used in research to investigate the reaction mechanism and chemical kinetics which always play a crucial role in selecting the best process from a number of competing options.

Similar techniques are used in the process development, the semi-technical stage to facilitate the scale-up of experimental systems to full-scale plant. However, it is on a full-scale plant that the unique advantages of radioisotope techniques completely manifest themselves. Therefore, in pursuit of process optimization, radioisotopes are used to help refine the design of reaction vessels as well as to study the performance under dynamic conditions such as hydrodynamics in multiphase flow system for chemical industries (Charlton, 1989).

The primary objective of this work is to describe and review the numerous industrial and experimental applications of radioactive tracer based techniques and to exploit the numerous potential benefits of these well-developed and established techniques.

Radioisotopes Used as Industrial Radiotracers

In 1913, the first investigations with isotopes tracers were carried out by de Hevesy and Paneth who characterized the solubility of lead (Pb) salts such as lead chloride, lead nitrate, etc. by using one of the naturally occurring radioactive materials of lead as the isotope indicator. After their discovery of the induced radioactivity, de Hevesy and Chiewitz in 1935, synthesized phosphorus-32 and used this tracer in fundamental life science research. On the same calendar year, de Hevesy and co-workers also conducted an experiment on the activation analysis of rare earth material (Gregory, Jan-Olov, Jan, & Christian., 2013). Nevertheless, according to International Atomic Energy Agency technical reports, radioactive tracers were first applied to industrial problem solving around the middle of the last century after which their usage has been increased continuously. The application of radiotracer based techniques are used extensively throughout the world for troubleshooting and process optimization over a wide area of applications in industry (IAEA, 2008).

A number of advantageous characteristics of radiotracer techniques have been identified. Firstly, radiotracer techniques provide good temporal and spatial resolution in both velocity and volume fraction which are good for phase hold-up measurements. In addition, the techniques are capable of providing instantaneous measurements and are also able to quantify the turbulent and dynamic flow structures on probe opaque systems in which the dispersed phase volume fractions are high. Statistically, this method is repeatable, and reproducible results can be obtained in a short and finite time. Radiotracer techniques are amenable to systems automation, which can minimize human involvement in the data collection process, so human error could be reduced. The setup is cost-effective and manageable to ensure the safety of personnel involved in the investigation and experimentations. Other desirable characteristics of radiotracer techniques relate to their portability and applicability to industrial and scale-up units as well as pilot plants (Al-Dahhan, 2008).

Unlike radiotracer techniques, there is no single experimental technique able to satisfy all the above-mentioned characteristics . There is, therefore, constant research in this field towards the direction of long-term goal to achieve the above requirements and desirable characteristics for other experimental diagnostics and measurement techniques in chemical and petrochemical industries (Chaouki et al., 1997; IAEA, 2008).

A variety of optically based techniques has been developed for flow visualization and quantification in transparent systems. However, photons of visible light do not pass through light-blocking objects. Since multiphase flow systems are opaque, only high energy gamma ray can be used because they can penetrate the systems to provide the information about the phase, flow, distributions, and mixing in the industrial process. Usually, gamma emitters are used in nuclear gauge densitometry to indicate liquid levels or provide an estimation line average of phases hold-up. Radioactive isotopes are used to trace and track the phases and provide residence time distributions (RTD), mean residence time (MRT), flow rate measurement, leak detection, etc. Recently, full quantification of the density distribution via computer tomography and full Lagrangian description of the flow of particle tracking became possible with the help of high computational power and advanced data acquisition technology. Many radioisotopes such as americium-241, bromine-82, caesium-137, chromium-51, gold-198, hydrogen-3, iodine-131, krypton-85, krypton-79, lanthanum-140, mercury-197, mercury-203, molybdenum-99, silver-110m, strontium-90, technetium-99m and xenon-133, can be used in industrial applications, especially in chemical and petrochemical industries. (Broadhead, and Heady, 1962; Lane, Nuckolls & Railey, 1963; Brownell, Farvar, Gyorey & York, 1965; Pilgrim, 1978; Campbell, Mirza, Thomson & Webb, 1984; Collins and Archundia, 1984; Iller, Przybytniak & St. Golembiowski, 1984; Larry, 1984; Burgio, Capannesi, Ciavola & Sedda, 1995; Torres, Olivares, De La Rosa & Lima, 1999; Kenneth, Janick, & Mark, 2006; Suga, 2002; Varga, Szalóki, Gáncs & Marczona, 2002; Adam and Laplace, 2003; Bondareva et al., 2005; Ding, Lin, Logan, Benveniste & Carter, 2005; Catán et al., 2007; Guevara, Repin, Catán, Jaćimović & Horvat, 2007; Pramanik, Bhattacharyya & Chattopadhyay, 2007; Yelgaonkar, Jayakumar, Singh & Sharma, 2007; Abu–Khadra, Abdel–Sabour, Abdel-Fattah & Eissa, 2008; Al-Dahhan, 2008; Shehee, Martin & Nash, 2009; Sukhoruchkin and Soroko, 2009; Pant et al., 2009; Jung et al., 2010; Kasban et al., 2010; Stęgowski et al., 2010; Guo et al., 2011; Mumuni et al., 2011; Pant et al., 2011; Kumar, Pant, Sharma, Mohan & Mahajani, 2012; Koron, Bratkič, Guevara, Vahčič, Horvat, 2012; Rhodes, 2012; Singare, 2012; Ugur and Sahan,. 2012; Affum, Adu, Dagadu, Coleman & Addo, 2013; Sugiharto et al. 2013b; Assadnassab, 2014; Belov, Kulkarni, Sohn & Murch, 2014; Othman et al., 2014; Takata, Tagami, Aono & Uchida, 2014; Vieira, Eduardo, Brandão & Braz; Zych et al., 2014; Dwivedi, Pathak, Kumar, Tripathib & Bajaja, 2015). The most commonly used industrial radiotracers in the chemical process investigations are tabulated in Table. 1.

Applications of Radiotracer Techniques in the Chemical Industry
Numerous types of multiphase flow systems are currently being used in the chemical industry. These systenms include multiphase reactors, mixing reactors, blending reactors, and separators/centrifuges. Radiotracer techniques have been applied to these systems for troubleshooting, diagnosing, online monitoring, process optimization, and development of advanced scale-up models. Examples for two-phase and three-phase fluidized and slurry bubble columns, and trickle bed reactors are briefly reviewed here. Additional examples and results of these techniques used for other types of multiphase flow systems related to chemical and petrochemical processes are also presented. These include investigations in distillation columns, extraction columns, structured beds, fluidized beds, gas-solid riser, circulating beds, water flow rig, stirred tanks, leaching tanks, grinding mills and mechanical

mixing equipment. Summary of the radiotracer application from several literature studies reviewed are tabulated in Table. 2.

A few radioisotope based techniques have been developed and facilitated for research at pilot plant scales, laboratory scales, industrial scales, and for site applications in industrial processes. These techniques are divided into three categories namely RTD measurement, sealed source for gamma-ray and x-ray transmission measurements, and radioactive and positron emission particle tracking.

Table 1: Most commonly used industrial radiotracers in chemical process investigations.

Isotope	Nuclear Reaction	Radiation and energy (MeV)		Half-life	Chemical form	Tracing of Phase	Reference
Americium-241	$^{240}Am(n,\gamma)\,^{241}Am$	Alpha: Gamma:	5.48(85%) 0.059(38%)	432 y	Americium Chloride Americium Citrate	aqueous solids	Bondareva et al., 2005; Shehee et al., 2009;
Argon-41	$^{40}Ar\,(n,\gamma)\,^{41}Ar$	Gamma:	1.29(99%)	1.83 h	Argon	gases	Colyar, Kressmann, Boyer, Schweitzer & Viguie, 2000; Yelgaonkar et al., 2007
Bromine-82	$^{81}Br\,(n,\gamma)\,^{82}Br$	Gamma:	0.55(70%) 1.32(27%)	36 h	Ammonium Bromide P-Dibromobenzene Dibromobiphenyl	aqueous organic organic	Kasban et al., 2010; Pant et al., 2011; Kumar et al., 2012; Singare et al., 2012; Othman et al., 2014
Caesium-137	$^{136}Cs\,(n,\gamma)\,^{137}Cs$	Beta: Gamma:	0.51(95%) 0.66(85%)	30.1 y	Caesium Chloride	solids (absorbed)	Abu–Khadra et al., 2008; Ugur et al., 2012; Zych et al., 2014; Dwivedi et al., 2015
Cobalt-60	$^{59}Co\,(n,\gamma)\,^{60}Co$	Beta: Gamma:	0.31(99.8%) 1.17(99.8%) 1.33(100%)	5.27 y	Cobalt Sulfine Cobalt Metal	solids (particles)	Kolics et al., 1992; Adam et al., 2003; Yelgaonkar et al., 2009
Chromium-51	$^{50}Cr\,(n,\gamma)\,^{51}Cr$	Gamma:	0.320 (9.8%)	28 d	Cr-EDTA Chromium(III) Chloride	aqueous	Pilgrim et al., 1978; Collins et al., 1984; Varga et al., 2002
Gold-198	$^{197}Au\,(n,\gamma)\,^{198}Au$	Gamma:	0.41 (99%)	2.7 d	Chloroauric Acid	solids (absorbed)	Al-Dahhan, 2008; Jung et al., 2010; Stęgowski et al., 2010; Vieira et al., 2014; Mumuni et al., 2011
Hydrogen-3	$^{2}H\,(p,\gamma)\,^{3}H$	Beta:	0.018 (100%)	12.6 y	Tritiated Water	aqueous	Campbell et al., 1984; Ding et al., 2005; Rhodes et al., 2012
Iodine-131	$^{130}I\,(n,\gamma)\,^{131}I$	Gamma:	0.36(80%) 0.64(9%)	8.04 d	Potassium or Sodium Iodide Iodobenzene	aqueous organic	Kasban et al., 2010; Stęgowski et al., 2010; Sugiharto et al., 2013
Krypton-85	$^{84}Kr\,(n,\gamma)\,^{85}Kr$	Gamma:	0.51(0.7%)	10.6 y	Krypton	gases	Brownell et al., 1965; Larry et al., 1984
Krypton-79	$^{78}Kr\,(n,\gamma)\,^{79}Kr$	Gamma:	0.51(15%)	35 h	Krypton	gases	Iller et al., 1984;

							Yelgaonkar et al., 2007
Lanthanum-140	$^{139}La\ (n,\gamma)\ ^{140}La$	Gamma:	1.16(95%) 0.92(10%) 0.82(27%)	40 h	Lanthanum Chloride	solids (absorbed)	Lane et al., 1963; Pant et al., 2009; Affum et al., 2013
Mercury-197	$^{196}Hg\ (n,\gamma)\ ^{197}Hg$	Gamma:	0.077(19%)	2.7 d	Mercury Metal	mercury	Guevara et al., 2007; Catán et al., 2007; Koron et al., 2012
Mercury-203	$^{202}Hg\ (n,\gamma)\ ^{203}Hg$	Gamma:	0.28 (86%)	46.6 d	Mercury Metal	mercury	Torres et al., 1999; Pramanik et al., 2007
Molybdenum-99	$^{98}Mo\ (n,\gamma)\ ^{99}Mo$	Gamma:	0.18(4.5%) 0.74(10%) 0.78(4%)	67 h	Sodium Molybdate	aqueous	Broadhead et al., 1962; Kenneth et al., 2006; Kasban et al., 2010
Selenium-75	$^{74}Se\ (n,\gamma)\ ^{75}Se$	Gamma	0.12(17%) 0.14(58%) 0.26(58%)	119.8 d	Ammonium Selenite	aqueous	Li et al., 2001; Jacimovicet al., 2009; Kühbacheret al., 2009
Silver-110m	$^{109}Ag(n,\gamma)\ ^{110m}Ag$	Beta:	1.50(13%) 1.47(4.0%) 1.38(24%)	249.8 d	ArgentumNitrate	solids (absorbed)	Adam et al., 2003; Sukhoruchkin et al., 2009; Belova et al., 2014
Sodium-24	$^{23}Na\ (n,\gamma)\ ^{24}Na$	Gamma:	1.37(100%) 2.75(100%)	15 h	Sodium Carbonate	aqueous	Tugrul et al., 1994; Altinsoy et al., 1999; Pant et al., 2009; Baytaş et al., 2013
Strontium-90	$^{89}Sr\ (n,\gamma)\ ^{90}Sr$	Beta:	0.546 (100%)	29 y	Strontium Chloride Strontium Titanate	aqueous	Abu–Khadra et al., 2008; Takata et al., 2014;
Scandium-46	$^{45}Sc\ (n,\gamma)\ ^{46}Sc$	Gamma:	0.89(100%) 1.84(100%)	84 d	Scandium Oxide	solids (particles)	Williams and McCarthy, 1987; Yunos, Hussain, Yusoff & Abdullah, 2014; Goswami et al. 2014
Technetium-99m	$^{99}Tc\ (\gamma,\gamma')\ ^{99m}Tc$	Gamma:	0.14(90%)	6 h	Sodium Pertechtenate	aqueous	Guo et al., 2011; Othman et al., 2014; Assadnassab et al., 2014
Xenon-133	$^{132}Xe\ (n,\gamma)\ ^{133}Xe$	Gamma: Beta:	0.08 (38%) 0.03(7%) 0.10(99%)	5.27 d	Xenon	gases	Suga et al., 2002; Burgio et al., 1995;

Table 2: Summary of Radiotracer Applications

Investigator	Radioisotope	Activity	System	Phase	Operating conditions	Parameters investigated
Thereska, Dida, Plasari & Cuci, 1991	Krypton-85	1 GBq	SO_2-Oxidation industrial reactor	Sulfur dioxide (Gas)	Four catalytic beds and three internal intermediate heat ex-changers with total height of the reactor is 13.0 m.	Gas flow distribution Residence time distribution
Burgio et al., 1995	Xenon-133	100 MBq	Coke pilot oven	Distillation gas (Gas)	200 000 m^3/h exhaust stream of a blast furnace.	Leak detection
Pant et al., 2000	Technetium-99m	10–20 MBq	Trickle bed reactor (TBR)	Glass beads and air (Gas-solid)	Liquid and gas flow rates used were 0.83×10^{-7}–16.67×10^{-7} m^3/s and 0–3.33×10^{-4} m^3/s	Residence time distribution Mean residence time Liquid holdup
Rammohan et al., 2001	Scandium-46	80 µCi	Stirred tanks reactors	Water (Liquid)	16 scintillation detectors; diameter vessel is 0.2 m; Rushton turbine agitator six rectangular blades	Radioactive particle tracking (RPT) Measuring the flow field Evaluate of dead and active zones
Degaleesan et al., 2002	Scandium-46	250 µCi	Air-water bubble column	(Gas-liquid)	Operated at different superficial gas velocities 2.4 and 9.6 cm/s, respectively, in a 14 cm diameter column	Measure the liquid velocity field
Tugrul et al., 2002	Sodium-24	0.4 MBq	Open channel test rig	Water (Liquid)	The open channel was filled in by means of three valves and 10 cm height of water was obtained	Flow measurement in open channels
Luo et al., 2003	Scandium-46	250 µCi	Photobioreactors (PBR)	Water and air (Gas-liquid)	Different type of spargers; Water at room temperature and pressure at superficial gas velocities (U_g) of 1 and 5 cm/s	Measure the flow field Irradiance patterns Controlling step of biomass productivity
Bhusarapu et al., 2004	Scandium-46	-NA-	Solids - risers of circulating fluidized beds	Glass beads and air (Gas-solid)	Superficial gas velocities from 3.2 to 4.5m/s and two different solids loading of 140 and 190 lbs at ambient pressure and temperature	Residence time distribution Solids circulation rates Solids flow pattern
Khopkar et al., 2005	Scandium-46	100 µCi	Gas–liquid stirred reactors	(Gas–liquid)	Two volumetric gas flow rates of 0.042 and 0.084. Flow regimes 3–3 cavity and flooding (ragged cavity) regimes	Circulation time distribution Gas–liquid flow identifications Radioactive particle tracking
Rados et al., 2005	Scandium-46	50 µCi	Slurry bubble column reactors	Water, glass beads and air (gas–liquid–	Working pressure of 1.45MPa and a maximum corresponding flow rate of 8.8m^3/ min	Radioactive particle tracking Liquid/slurry circulation rate

				solid)		
Fraguio et al., 2006	Scandium-46	100-200 µCi	Three-phase fluidized beds	Water, glass beads and air (gas–liquid–solid)	Gas superficial velocities were varied within the range $0<\mu_G<0.11$ m/s. Liquid superficial velocities of 0.058 m/s for PVC particles and 0.065 m/s for the glass beads	Characterize the motion of the solid Radioactive particle tracking Measurement of flow regime
Klusener et al., 2007	Argon-41	-NA-	Horizontal bubble column reactors	Ethylbenzene (Gas-Liquid)	reactors with a very low aspect ratio: height 4–6 m, length 15–25 m	Residence time distribution Backmixing measurements
Doucet et al., 2008	Scandium-46	125 µCi	Patterson–Kelley V-blender	Glass beads (Solid)	3-mm glass beads (2.5 kg/L) fill level was 40% in volume. Blender ensured coupled to a 50:1 gear reducer	Flow of particles mapping Radioactive particle tracking Measuring mean velocity field
Sugiharto et al., 2009	Iodine-131	-NA-	Hydrocarbon transport (HCT) pipeline	Water, crude oil and gas (Gas-liquid)	Consisting of water 95%; crude oil 3%; and gas 2%, at temperature of $70^{\circ}C$	Separation length Residence time distribution
Pant et al., 2009	Gold-198	25 MBq	Circulating fluidized bed system (CFBS)	Coal particles and air (Gas-solid)	Bed height 0 – 1.5mm. Fluidization velocity 1.11-1.5m/s Loop seal velocity 0.08 – 0.27m/s	Circulation time Mean residence time Residence time distribution
Din et al., 2010	Technetium-99m	0.5 mCi	Pulsed sieve plate extraction column	Water and kerosene (Liquid-liquid)	Dispersed and continuous phase superficial velocity ($U_d = 0.34\times10^{-2}$ m/s) and ($U_c = 0.37\times10^{-2}$m/s)	Residence time distribution Evaluate holdup of dispersed phase
Kasban et al., 2010	Molybdenum-99	300µCi	Water flow rig	Water (Liquid)	Tank with a diameter of 30cm and a height of 80cm. The tank contains four stirrers; each is 20cm in length	Residence time distribution Mixing time and the flow rate
Stęgowski et al., 2010	Iodine-131	1.8 Ci	Leaching tanks	Gold particles and cyanide (Liquid-solid)	Continuously stirred tank at rotational speed of 960 rpm by a mechanical agitator	Mean residence time Flow rate measurement Volumes of the mixing zones
Abdelouahed et al., 2011	Technetium-99m Iodine-131	150 mCi 50 to 250 mCi	Continuous stirred tanks of plug flow type	Phosphoric acid (Liquid)	Ranged from about 0.2 kW/m^3 for moderate mixing to 2 kW/m^3 for intense mixing	Residence time distribution Measurement of flow rates Mixing efficiencies
Mumuni et al., 2011	Gold-198	40 mCi	Clinker Grinding Mills	Cement powder (Solid)	Two chambers with different sizes of grinding balls rotates at a speed of 16 rpm	Residence time distribution Mean residence time
Kumar et al., 2012	Bromine-82	10 MBq	Trickle bed reactor (TBR)	H$_2$ gas and Water (Gas-liquid)	Liquid and gas flow rates ranged from 0.83×10^{-7}–16.67×10^{-7} m^3/s and 0–3.33×10^{-4} m^3/s	Mean residence time Holdup of liquid phase Residence time distribution

Pant et al., 2012	Bromine-82	20–40 MBq	Rotary fluidized bioreactor (RFBR)	Wastewater and air (Gas-liquid)	Wastewater flow rates ranging from 7.5 to 12.5 m^3/day and air flow rate ranges from 9 to 15 m^3/h	Residence time distribution The degree of mixing
Widatalla et al., 2012	Technetium-99m	1 mCi	Closed circuit water flow rig	Water (Liquid)	Tank of 175L with four stirrers with flow rate of water ranged from 5-15 L/min	Residence time distribution Bypass volumetric rate Tank dead volume
Sugiharto et al., 2013	Bromine-82	1 mCi	Water tank pipeline	Water (Liquid)	Horizontal stainless steel pipeline of 3" (7.62x10^{-2} m) inner diameter with capacity of 2,500 l.	Flow rate measurement
Goswami et al., 2014	Gold-198	11 MBq	Fluidized bed	Sand particles and air (Gas-solid)	Mixing times at static bed heights 0.3, 0.45 and 0.8 m and gas velocities from 0.1 m/s- 0.6 m/s	Mixing times and solids holdup
Othman et al., 2014	Technetium-99m	10 μCi	Integrated continuous mixing flow rig	Water (Liquid)	100 rpm impeller speed, 50mm impeller clearance, Type A mixer, and 900 s sampling time	Residence time distribution Mean residence time Percentage of dead volume
Pant et al., 2014	Iodine-131	55.5 GBq	Branch canal dam	Water (Liquid)	Concrete Volute Pumps capacity of 20 m^3/s and Vertical Turbine Pumps (VTPs) capacity of 5 m^3/s	Discharge rate measurement
Samantray et al., 2014	Bromine-82	55 mCi	Diesel hydrotreater (DHDT)	Diesel (liquid)	The pressure exchanger 130 to 135 kg/cm^2 pressure in the tube side of the exchanger 105 to 110 kg/cm^2. The temperature heat exchanger 300 to 350ºC	Leak detection in a heat exchanger system
Zych et al., 2014	Caesium-137	70 and 100 mCi	Sedimentary suspension flow rig	Water and sand (Liquid-solid)	Various depths of the trough (40, 80 and 160 mm) for a similar carrying liquid velocity	Measurements of slurry flow rate Model of compound two-phase flow verification
Pant et al., 2015	Bromine-82	40–60 MBq	Pilot-scale continuous leaching reactor	Water (Liquid)	The volume of each compartment is 260L, 175L and150L. Water as flowing phase and at ambient conditions.	Mean residence time Dead volume measurement Residence time distribution

Table 3: Radioisotope based technique investigations reported by Imperial Chemical Industries (ICI) company in one year (Charlton, 1986).

Technique	Number of applications
Level and interface measurements:	
Gamma-ray transmission absorption	210
Neutron backscattered	480
Gamma-ray backscattered	71
Blockage detection and deposition:	
Gamma-ray transmission absorption	132
Neutron backscattered	129
Entrainment and voidage:	
Gamma-ray absorption	86
Thickness and corrosion measurements	15
Distillation-column scans	108
Flow measurements:	
Pulse velocity	483
Dilution techniques	84
Leak detection	90
Residence-time distributions studies	21
Carryover studies (radiotracer)	6

The list is not fully comprehensive as less commonly used techniques have not been included.

Various types of photons transmission measurement techniques by radioisotopes have been developed and could be used for industrial investigations and research such as level monitoring and control using nuclear gauge densitometry, difference average densities (phase holdups) using gamma-ray densitometry, time and cross-sectional averaged phases hold-up distribution along the reactors or equipment height using computed tomography (single energy, dual or multiple energy), single photon emission computed tomography, positron emission tomography and Compton scattering tomography using different types of radioisotopes (Al-Dahhan, 2008). For the other categories, a single radioactive particle or multiple radioactive particles usually made from solid material have been used for radioactive particle tracking (RPT) techniques. These techniques have been used to estimate three-dimensional flow fields, calculate phase trajectories, determine many flow characteristics including velocities, RTDs, turbulent parameters, stagnant zones, diffusivity, turbulent kinetic energy using various types of radioactive tracers in aqueous or solid forms (Lin, Chen, Chao, 1985; Larachi, Kennedy & Chaouki, 1994; Doucet et al., 2008; Khanna, 2008 Dubé, Chaouki & Bertrand, 2014).

There are numerous ways to utilize industrial radiotracers in a chemical and petrochemical industry. A large number of manufacturing companies around the world employ radioisotope based techniques for production of metals, chemicals, plastics, pharmaceuticals, paper, rubber, clay and glass products, food, tobacco, textiles, and many other related products. Thus, the radioisotope based techniques are used to study the mixing efficiency, the effect of chamber geometry, RTD in reactors, and flow rates and patterns measurements in columns and towers for process investigations and optimizations. and few are sampled in Table.3. Some of the techniques described below indicate the scope and number of the industrial applications of radioisotopes in a year reported by Imperial Chemical Industries (ICI) Company.

Design of Radiotracer Technology Experiments

The literature on methods for troubleshooting and process optimizations as well as model development in radiotracer technology experiments are growing rapidly. The two techniques act as the sole problem solver; among these are the tracer impulse response. Its application is to discover the RTDs, dispersion extent and dispersion coefficient, phases hold-up, or to match the response to a model; whereas gamma-ray densitometry is used to acquire the line of average holdups and from a number of measurements it determines radial profiles and assesses the potential problem areas within the examined instruments. On top of that, these techniques are very valuable and advancement has been made in the technology used for their applications. Due to the advancement in technology and computational approach, several contemporary techniques have been developed and utilized as indicated above. Among these, the design of experimental RTD and RPT techniques are discussed here. Furthermore, their crucial applications on multiphase flow systems employed in chemical and petrochemical process are briefly demonstrated.

There are several issues in practical considerations before the implementation of radiotracer techniques. These factors include the availability of the radioisotope for the element to be traced with proper characteristics such as half-life, energy types, radioactivity and tracking phase. Calculation of the amount (quantity and activity) of the radiotracer for the experiment is required, and is a function of the system (volume) to be studied and its half-life. This activity consideration is also important from a radiological and safety point of view. However, there are situations where high level (millicuries) of gamma-ray emitters are utilized. For instance, 10 mCi of ^{24}Na will return a dose of about 204 milliroentgens per hour (at 1ft distance) (Ashfaq, 2007). One should also be quite cautious about the radiation dose delivered to the hands and fingers while handling radioactive materials. Lastly, full attention must also be given to the disposal of radioactive wastes resulting from the experiment and the possible disposal methods will depend on the specific radioisotope availability, concentration and activity, and the characteristics of the waste. Due to the opaque nature of many systems, especially in the case of multiphase flows, two methods based on radioactivity have been considered in the application of RPT and RTD in the design of radiotracer experiments (Pant, 2001; Al-Dahhan, 2008; Othman et al., 2014)

Radioactive Particle Tracking (RPT)

In RPT, the trajectory of a radiotracer particle is designed to be the marker of the phase whose velocity is to be mapped and tracked by a scintillation detector. Figure 1 shows the RPT setup consisting of a radioactive tracer particle, radiation detectors, data acquisition system and a calibration device. The radioactive tracer particle is normally a gamma radiation emitter of high energy γ-rays with an activity of 50-500 μCi such as ^{198}Au, ^{137}Cs and ^{46}Sc (Upadhyay and Roy, 2010; Vieira et al., 2014). The particle is intentionally made to be neutrally buoyant to track liquid phase and solid particles. The dimension, form, appearance and density of the tracer particle is matched to the material of the solid particles and is dynamically similar to the particulate phase. Usually, the radioactive particle is composite prepared by inserting a certain mass of radioisotope into hollow polypropylene particle beads of diameter 0.7 mm to 2.4mm. An array of scintillation detector at a sampling frequency up to 600 Hz is mounted on the vessel or column to track the movement of the tracer and record the photon counts. Moreover, the position of the tracer particle with respect to time can be investigated by determining the amount of radiation sensed by each detector. A 3-D velocity profile of the tracer particles and diversification of the particulate phase in the hydrodynamic process can be easily generated by tracking the tracer movement over time (Drake et al., 2011).
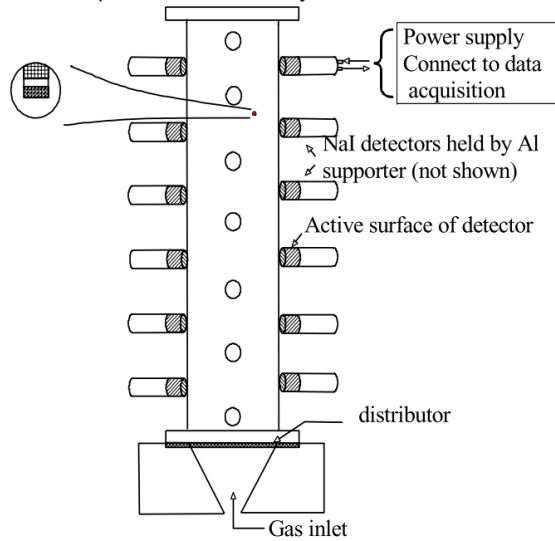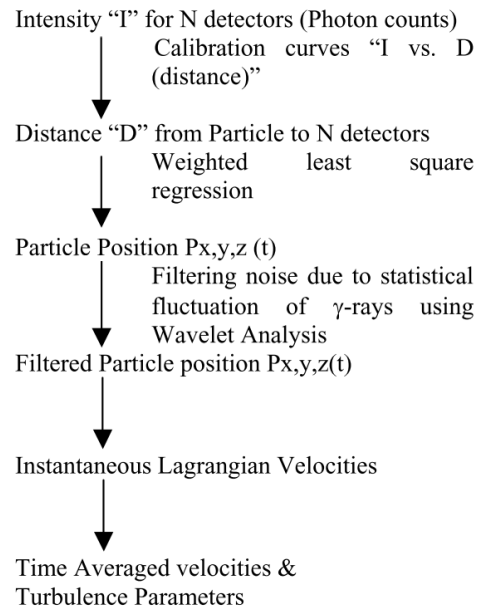
Figure 1: Schematic Diagram of the RPT Facility at the Chemical Reaction Engineering Laboratory (CREL)-Washington University (WU) (Luo, 2005)

Lin et al. (1985) discovered the first modern RPT set up to investigate the recirculation pattern of solid particles in circulating bubble fluidized beds. The reconstruction of tracer position inside the vessel was obtained by curve fitting the calibration data from the polynomial functions based on the principle that the number of γ-rays counted by the detector is dependent solely on the distance between the point tracer and the virtual centre of the detector (Khanna, 2008). The system was calibrated by mounting the radiotracer particle at necessary locations inside the vessel where the polynomial function was utilized for photon counts collection. Lin et al. (1985) succeeded in improving the upgraded version of the RPT system at Chemical Reaction Engineering Laboratory (CREL) at Washington University in St. Louis and was later used to study gas-liquid and gas-solid fluidized beds, packed beds, bubble columns, high pressure and high temperature bubble columns, slurry bubble columns, and liquid-solid circulating fluidized bed risers (Devanathan and Dudukovic 1990; Dudukovic, Bhusarapu & Al-Dahhan, 2006; Kumar et al., 1994; Yang et al., 1992). However, Larachi et al. 1994 discovered a mathematical phenomenological model that relates the interaction of photons with the vessel geometry to determine the position of the tracer particle from the data counted by the scintillation detectors. A map of counted data by scintillation detectors for a large number of positions inside the vessel was created using Monte-Carlo simulation. The simulation data was improved by a calibration process during which a tracer particle was placed at specified locations. The data counted by the scintillation detectors were then compared with the simulated data from Monte-Carlo to compute the optimal model parameters. The number of calibration points needed were considerably smaller compared to the work done by Lin et al. (1985) . Moreover, Godfroy et al. (1997) developed neural networks that manage to reduce computational time to reconstruct the position of tracer particle as compared to the works done by Lin et al. (1985) and Larachi et al. (1997). Nevertheless, a large number of data points need to be calibrated before the neural network could determine the position of the tracer particle.

Vieira et al. (2014) carried out RPT experiments with a new tracing methodology which relates the trajectory of a particle tracer moving in a fluid as a series of small cubic cells engaged by consecutive particle positions. A 2.8 mm final particle diameter of polyurethane coated with radioisotope ^{198}Au with 0.5mm radius spherical particle was prepared to achieve the nearest density of 1.001 kg/m^3. The result was a naturally buoyant particle close to the water density. The RPT setup for this experiments comprised of four laterally shielded 5.08 cm x 5.08 cm NaI:Tl detectors with RING geometry placed around a cylindrical PVC stirred tank reactor as shown in Figure 2. This research team finally discovered very useful information for calculating the coordinates and velocities of the particle at that time. According to their results, the maximum deviation found between theoretical and experimental values of the average rotating period was less than 8% with the ability to track the movement of the particle. With a maximum relative error equal to 7.8%, the particle's trajectories were successfully reconstructed improving its real movement (Vieira et al. 2014).
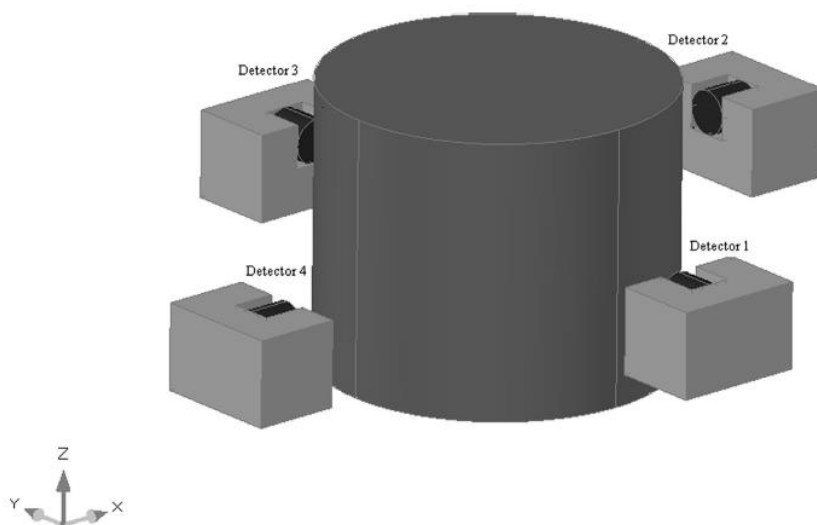
Figure 2: Design of the RING geometry with four detectors were mounted around a cylindrical PVC stirred tank reactor (Vieira et al. 2014).

The next case study was conducted by Upadhyay et al. (2010). The authors investigated the effect of air inlet velocity and bed composition of binary fluidized beds of particles of the same size and different densities on mixing and hydrodynamic behaviour using RPT technique. The RPT experiments were performed in 11.5 cm ID Perspex® cylindrical fluidized bed with a packed bed distributor of 2 cm in size individually for both solids by tracking one particle at a time. A mixture of sago particles (density 1350 kg/m3) and glass beads (density 2600 kg/m3) were used in the process where two gamma-ray emitters introduced as the radiotracers were Ceasium-137 (3 mCi) and Scandium-46 (3.2 mCi) which produce different energies; Cs-137 produces 662 keV (one photopeak energy) and Sc-46 produces 889 keV and 1120 keV (two photopeak energies). Interesting findings were discovered where significant occurrences of strenuous (jetsam) phase were seen to be fluidized by the action of the lighter (flotsam) phase even below its minimum fluidization velocity. Moreover, the increment of air inlet velocity resulted in the extensive mixing of the solid phases although the jetsam phase was not completely mixed at very high air velocities. An axisymmetric view of the bed is shown in Figure 3 which indicates that the solids ascended near the wall and descended near the centre at comparatively low fluidization velocity and low initial bed height in a time-averaged detection (Upadhyay et al., 2010).
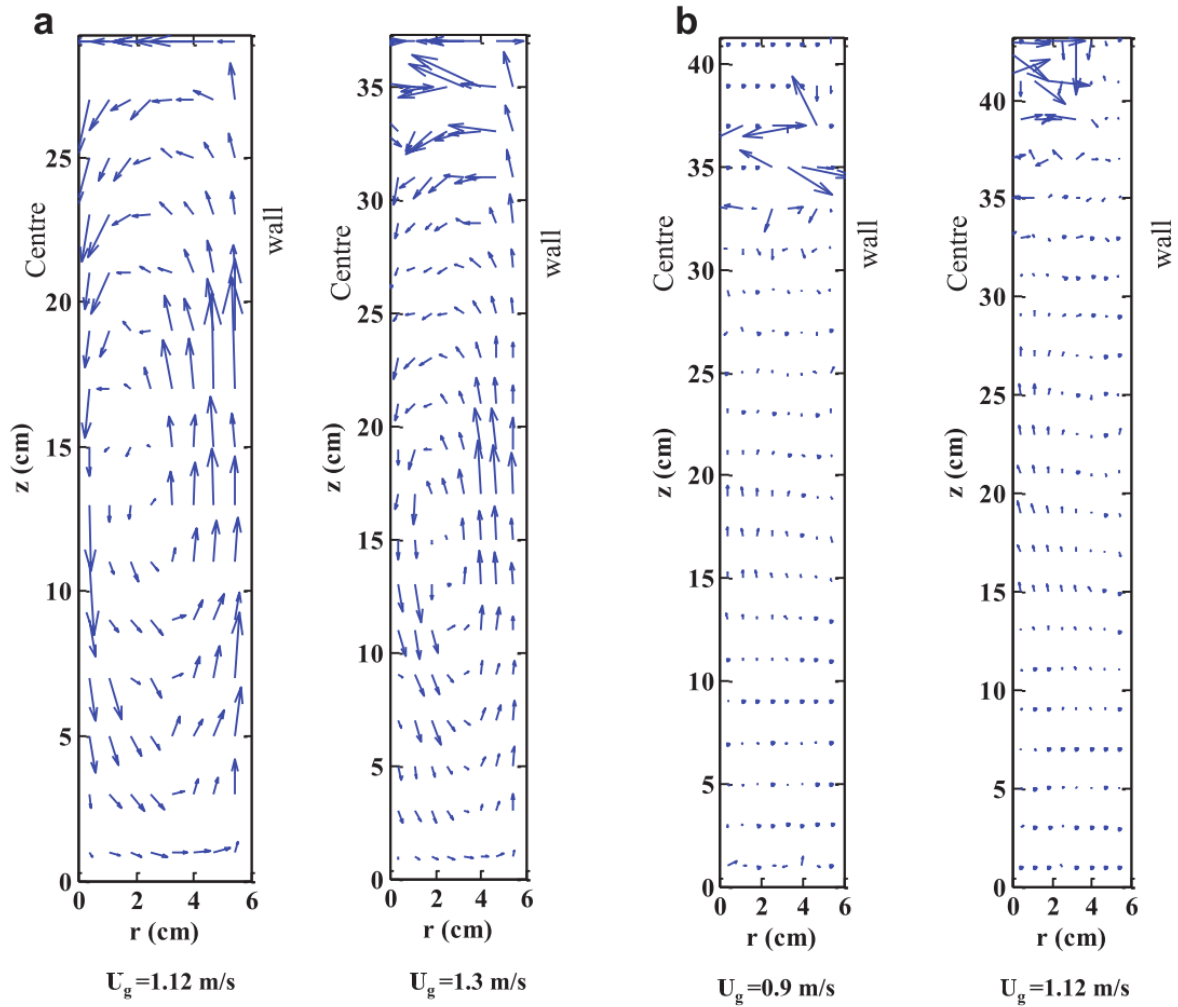
Figure 3. a: Mean velocity vector plot of pure glass for different inlet air velocity. b: Mean velocity vector plot of pure sago for different inlet air velocity (Upadhyay et al., 2010).
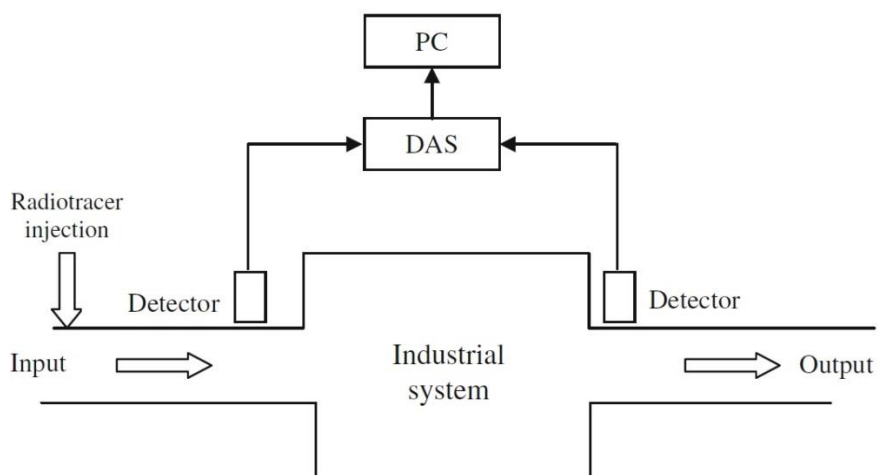

Figure 4: RTD measurements in an industrial system (Kasban et al., 2014).

Residence Time Distribution (RTD)

Generally, RTD measurement is done by injecting a suitable radiotracer at the system input and monitoring the concentration of the tracer using radiation detectors placed at the output of the system or taking samples from the system output at regular intervals as shown in Figure 4. A data acquisition system (DAS) is used to collect the signals from the detectors and a personal computer (PC) is connected to the DAS by coaxial cables. The signals from the detectors are collected by DAS and shown on the PC screen. An example of a measured RTD signal is shown in Figure 5.
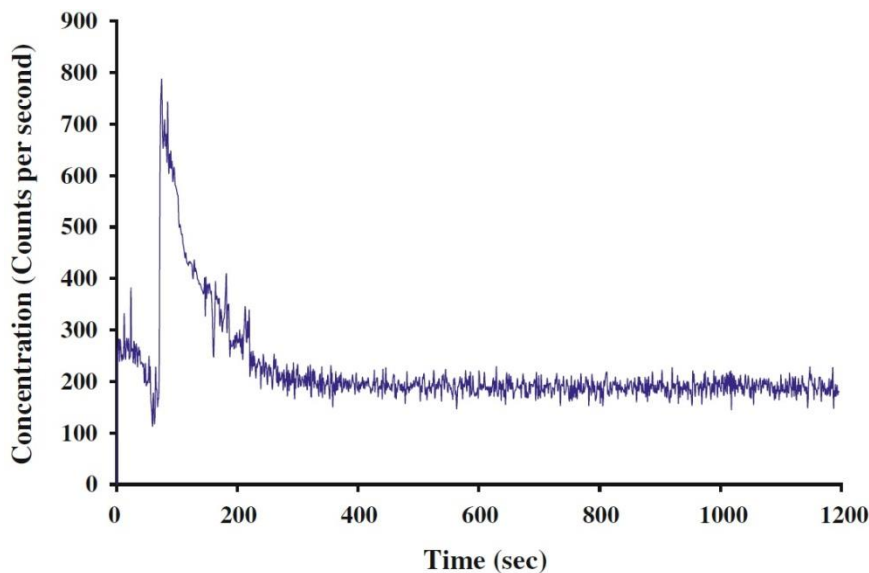
Figure 5: Example of measured RTD signal (Kasban et al., 2014)

RTD of liquid phase investigation using radiotracer ^{82}Br with 10 MBq activity was conducted by Kumar et al. (2012) in a trickle bed reactor (TBR). The reactor was filled with two different types of packing, hydrophobic and hydrophilic, one at each time, to investigate the dispersion characteristics of the liquid phase. The liquid and gas flow rates ranged from 0.83×10^{-7}-16.67×10^{-7} m^3/s and 0-3.33 $\times 10^{-4}$ m^3/s, respectively. An axial dispersion with exchange model was used to simulate the measurement of RTD curves and model parameters (Peclet number and MRT) to prove that the dispersion of the liquid phase is higher with the hydrophobic packing, whereas hold-up is higher in the case of hydrophilic packing.

The next case study was carried out by Pareek et al. (2001) in which the authors used a commercial titania powder as the radiotracer in investigating RTD of solids in an 18 L pilot-scale three-phase annular reactor as a function of gas and liquid flow rates. The objective was to characterize the influence of hydrodynamics on solids mixing both in co-current and counter-current operation. They reported that RTD curves for the co-current operation showed a bimodal behavior in which the peaks decreased as gas flow rate decreased, while the RTD behavior for counter-current operation was unimodal and was therefore modelled as a series of stirred tanks with a recycle stream. They concluded that the associated mean squared error was in the range of 1-4% and there seemed to be an evolution of solids flow pattern within the reactor as demonstrated by the time-dependency of the intensity function.

An alkaline based continuous leaching process is used for uranium extraction from uranium ore. The process is carried out as a continuous leaching reactor (CLR) and is expected to behave as a continuously stirred tank reactor (CSTR) for the liquid phase. According to Pant et al. (2015), the

pilot-scale CLR used in a Technology Demonstration Pilot Plant (TDPP) was designed, installed, and operated; thus needs to be tested for its hydrodynamic behavior. Pant and co-workers (2015) carried out the radiotracer technique in CLR for RTD measurement of liquid phase with Bromine-82 as ammonium bromide in which approximately 40-60 MBq activity was used in each run to characterize the flow behavior of the reactor and validate its design. The measured RTD curves were treated and mean residence times were determined and simulated using tanks-in-series model. The result of the simulation indicated no flow abnormality and the reactor behaved as an ideal CSTR for various operating conditions used in the investigation.

A series of radiotracer experiments were carried out by Othman et al. (2014) in order to determine the mean residence time (MRT) and percentage of dead zone, V_{dead} (%). The experiment was conducted in an integrated mixer consisting of Rushton and pitched blade turbine (PBT). The optimum conditions for the experiments were 100 rpm impeller speed, 50mm impeller clearance, Type A mixer and 900 s. Thus, a 4-factor 3-level Taguchi L9 orthogonal array was introduced to obtain an accurate optimization of mixing efficiency with minimal number of experiments. Moreover, ANOVA was performed to determine the most influential factors contributed in the study. The values of prob > F less than 0.05 indicated that all factors involved were significant. The R^2 value (0.95) for both outputs implied that 95% of the factors can be described well by the model. The predicted optimum levels of the control parameters for Type A mixer as well as an additional run for Type B mixer were validated by conducting an experiment at the conditions proposed by Othman et al. (2014).

The next case study demonstrated the application of radiotracer technology in measuring the RTD of wastewater in the rotary fluidized bioreactor RFBR. This study was conducted by Pant et al. (2012) using [82]Br as the radiotracer for its hydrodynamic behaviour to validate the design. The reactor was designed to operate with wastewater flow rates ranging from 7.5 to 12.5 m^3/day with predetermined quantity (20% of the reactor volume). The results of the investigation showed no flow abnormalities and the reactor behaved as an ideal continuously stirred-tank reactor at all operating conditions. This study reported that the design of the reactor was validated, thus the results of RTD analysis obtained in the present pilot-scale system could be used to scale-up the process.

Due to its extreme importance in various industrial and environmental applications, multiphase flow modelling becomes more challenging. In the present study, the prediction of separation length of multiphase flow was examined experimentally by the injection of two kinds of iodine-based radiotracer solutions into a hydrocarbon transport pipeline (HCT) with an inner diameter of 24 in. (60,96 m) consisting of 95% water, 3% crude oil and 2% gas. A series of radiotracer experiments conducted by Sugiharto et al. (2013b) used two radiotracers, I-13 and Na-24. This experiment was carried out in a pipe segment that was far from the branch points with the assumption that stratified flows in such segment have been achieved. Two detectors located at 80 m and 100 m from the injection point were used to investigate the RTD curve resulting from the injection of radiotracer solutions. The researchers concluded that the velocity of water flow was higher than the flow rate of crude oil in a water-dominated system despite the higher density of water and indicated that the placement of the first radiation detector at the distance of 80 m from the injection point was correct.

Moreover, Muroyama et al. (2013) determined the gas holdup, bubble size distribution, and Sauter mean diameter for oxygen micro-bubble dispersions in water using RTD in an acrylic acid resin column with an inner diameter of 0.15 m with a working liquid height varying from 0.500 to 1.850 m. The micro-bubbles which varied in their Sauter mean diameters (32 μm - 40 μm) depending on the gas velocity, were employed to measure their superior mass transfer properties which were enhanced

by the effects of self-compression and shrinking. Figure 6 shows a schematic diagram of the experimental apparatus used together with the RTD. The micro-bubble generator (AS-KS, ASP corporation) used in the experiment was a combination of high-speed rotation and compression-dissolution-type generators. Muroyama et al. (2013) also determined the increment in superficial gas velocity, and observed that the peak of the frequency distributions shifted towards larger diameters and the value of dvs increased slightly within the range of 32 μm to 40 μm.
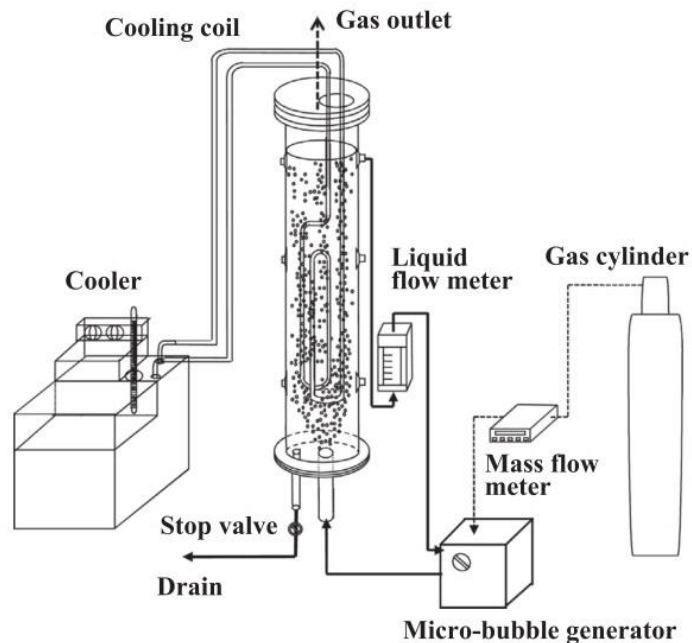
Figure 6: A schematic diagram of the RTD experimental apparatus
by (Muroyama et al., 2013)

Radiation Detection and Data Acquisition Technology

Currently, the development of radiotracer techniques for research at pilot plant scales, laboratory scales, industrial scales, and for site applications in industrial processes are becoming increasingly acceptable by industry players. In order to meet their demands, many multifarious mathematical models and reconstruction algorithms have been developed, advanced, tested and executed for image reconstructions. Furthermore, some of the mathematical methodologies, processed data, and randomized algorithms have been conducted from post-processing image reconstructions to rough approximations of desired parameters such as phase holdups, phase velocities, phase turbulent parameters and so on. Performing all these data acquisition techniques properly is a non-trivial mission. For instance, to execute RPT techniques, specialized devices and apparatus are needed as well as specialized knowledge and experience for particle preparation, data calibration, implementing the experiment, safe handling of the radioactive materials, data acquisition and data reconstruction, and post-processing skills. Chaouki et al. (1997) and IAEA reports (2008, 2006, 2005) have outlined and discussed some of these radiotracer techniques operating procedures and described the case studies conducted around the world.

Electronics and data acquisition systems are important components in radiation detectors that are to be applied to radiotracer pre-processing techniques. The performance of these systems has powerful implications on important parameters detection devices such as the sensitivity and the background noise. Currently, the detection technologies used in these data acquisition systems are scintillating crystals connected to photomultiplier sensors and semiconductor detectors. Hence, desired

characteristic of these detectors is the capability to initiate the generating pulse signal for each interacting particle.
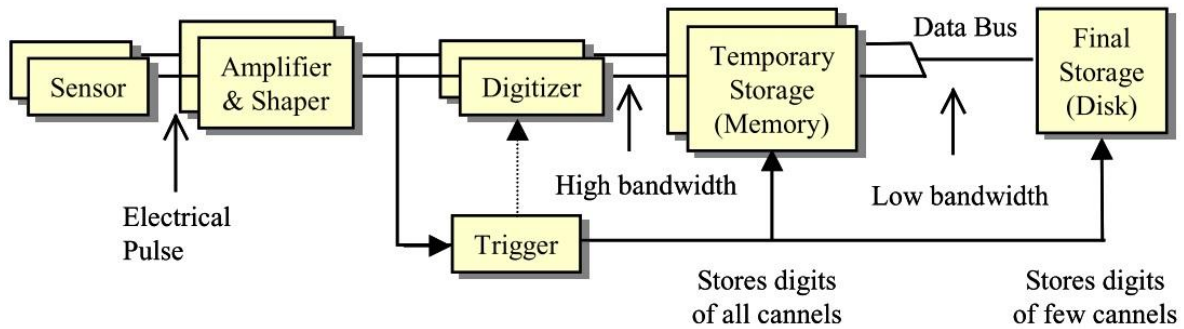

Figure 7: Generic block diagram of the data acquisition system of radiation detectors (Varela, 2004)

According to Varela (2004), the electronics and data acquisition systems are a part of the complex process that leads to the final processing data for analysis. The systems begin with the interaction of radiotracers emission from the tracked phases, followed by the physics processes involved in the detection of radiations and in the generation of corresponding electrical signals. The electronic systems are then responsible for analog processing of the detector electrical signals which basically amplify the detector pulses and convert the analog signal to digital outputs. The trigger system is responsible for identifying the existence of particle interactions in the detector (events) to set the stage for the data acquisition process. Furthermore, the read-out system will continue collecting digital data, selecting the flight statistics relevant to the designed. Finally these data are transferred to a data acquisition computer where the data are stored in a permanent medium (hard disk). Filed stored data contains a list of incidence for each record of the detected particles and its properties required.
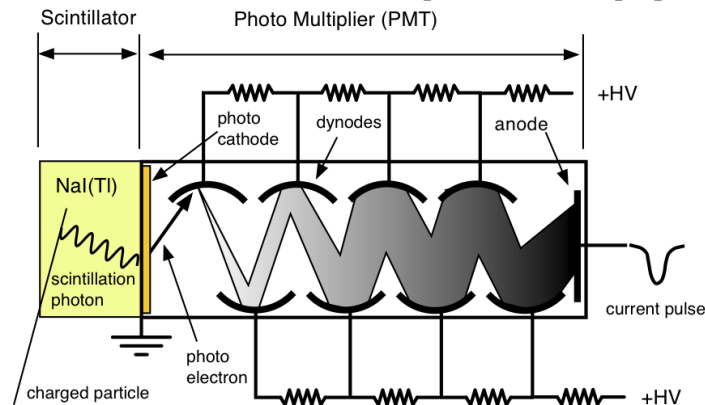

Figure 8: Schematic of scintillator and photo multiplier (PMT) (Werner, 2011).

Solid scintillation detectors are the most commonly used detectors based on a single crystal of thallium doped sodium iodide. It is also called NaI (Tl) detector. The crystal detector is optically coupled to a photomultiplier tubes (PMT) as illustrated in Figure 8. The interaction of a gamma photon with the scintillation crystal material evolves light emission, and then detected by the photomultiplier tubes. The principle of production of prompt and delayed scintillation light by incident radiation is illustrated in Figure 9. The light output has a constant ratio to the gamma energy. The electronic system integrated with the photomultiplier tubes scrutinize the pulses according to pulse amplitude (energy) and accumulate the results in a multichannel analyzer. Thus, the energy and intensity are recorded, and the result is the gamma energy spectrum of the radiation source. On the other hand, these scintillation NaI(Tl) detector has a high intrinsic efficiency but a limited energy

resolution. The scintillation crystals have different sizes; however the larger the crystal, the higher the price accordingly, and the efficiency for high gamma energies increases in line with the detector volume. Generally, common counting equipment has cylindrical crystal size of 5.08×5.08 cm^2 (2 in. $\times$ 2 in.) to 12.7×12.7 cm^2 (5 in. $\times$ 5 in.) (height $\times$ diameter). Roy, Larachi, Al-Dahhana & Dudukovic (2002) reported that smaller crystal has poor resolution but marginally high sensitivity. Furthermore, 2 in. $\times$ 2 in. nominal size crystal detector is recommended for high-resolution and good economics. Thus, a NaI detector with size of 2 in. $\times$ 2 in. was found to be more efficient. Figure 10 shows the effect of crystal detector size on sensitivity and resolution of the detector axis.
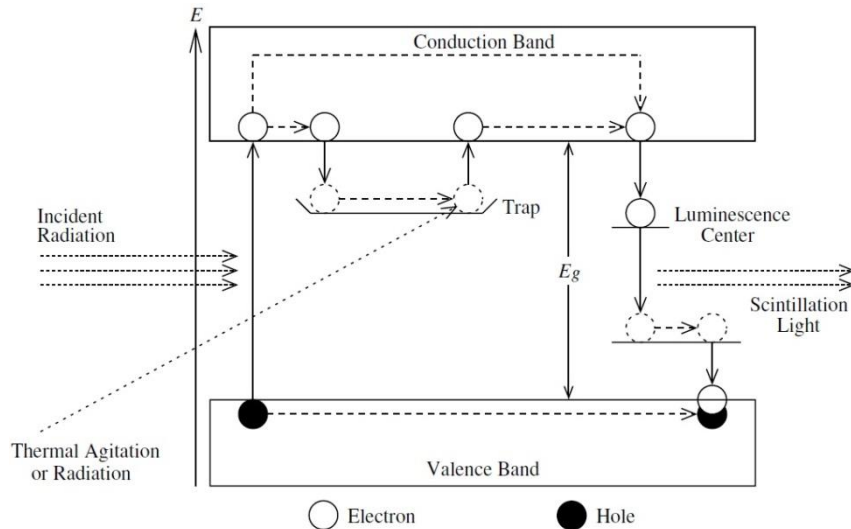

Figure 9: Principle of production of prompt and delayed scintillation light
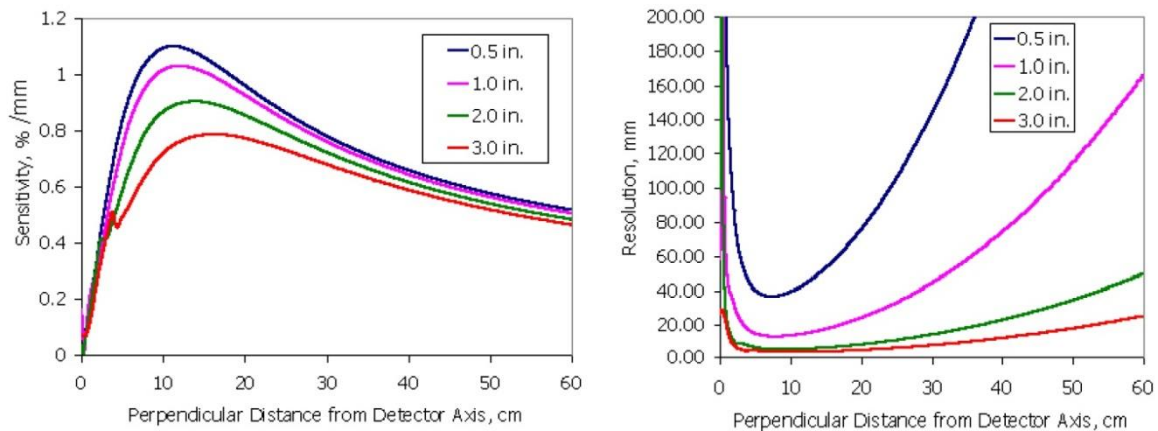by incident radiation. (Ahmed, 2014)

Figure 10: Effect detector crystal size on sensitivity and resolution from the
detector axis. (Roy et al., 2002)

In a large number of applications, one is interested in counting the number of interesting pulses. The height of a pulse depends on the energy deposited by the radiation and the expected energy is in the range of interest. Thus, the remaining pulses must be filtered or blocked to obtain a clean dataset. Since there are always low-level noise and high-level spurious pulses, the filtration requires two-level discriminator that discriminates the pulses based on the thresholds set and produces logic outputs known as single channel analyzer as shown in Figure 11. Here ULD and LLD stand for upper-level discriminator and lower-level discriminator respectively. As illustrated in Figure 12, multichannel analyzers have a number of channels such as 512, 1024, or more, provide the user some choices in selecting the appropriate resolution. Most multichannel analyzers can be operated in two different

modes which are the usual pulse height analysis mode and scalar mode. In this mode, the pulses are counted with respect to some other parameters of interest. It could even be time, in the case where the system simply measures the total intensity.
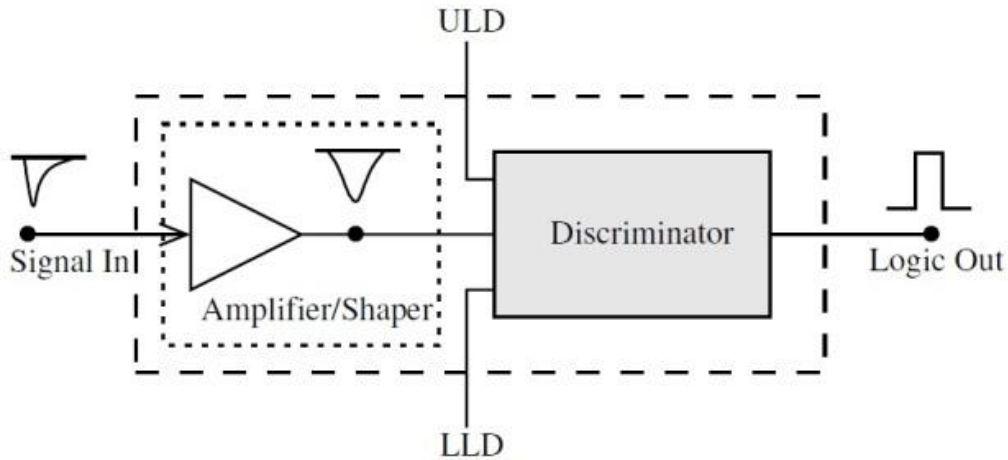

Figure 11: Block diagram of a simple single channel analyzer. (Ahmed, 2014)
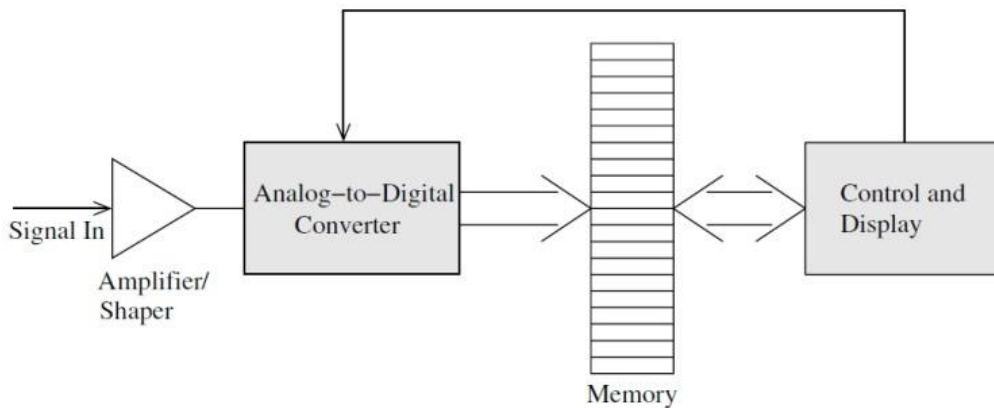

Figure 12: Block diagram of a simple multi-channel analyzer designed for pulse height analysis. (Ahmed, 2014)

Minimum of two radiation detectors are required for simple radiotracer experiments such as measurement of the RTD of the radiotracer inside pipelines (inlet-outlet response) and flow rate measurement or leak detection in a simple or laboratory scale reactors. Meanwhile several detectors (4-6 units) are required to obtain information in certain sites of the processing vessels or wastewater reactors, and as many as possible (> 10-20) are necessary for complex engineering installations like solid-liquid risers, photobioreactor, fluid catalytic cracking units (FCCU) or for radioactive particle tracking and tomographic measurements. Data acquisition system which collects signals from the radiation detectors is the important equipment for online radiotracer to ensure the visualization of the data. The dead time between two simultaneous measurements is normally less than 1 μs and the minimal dwelling time is 1-2 ms to ensure the visualization of data could be close as possible to the real time experiment. Sodium iodide (NaI) scintillation detectors are strategically placed on the system of interest which is connected to amplifiers/discriminators for signal adjustments and the latter are connected to high-speed counter cards which transfer the information to data acquisition system as illustrated in Figure 13.
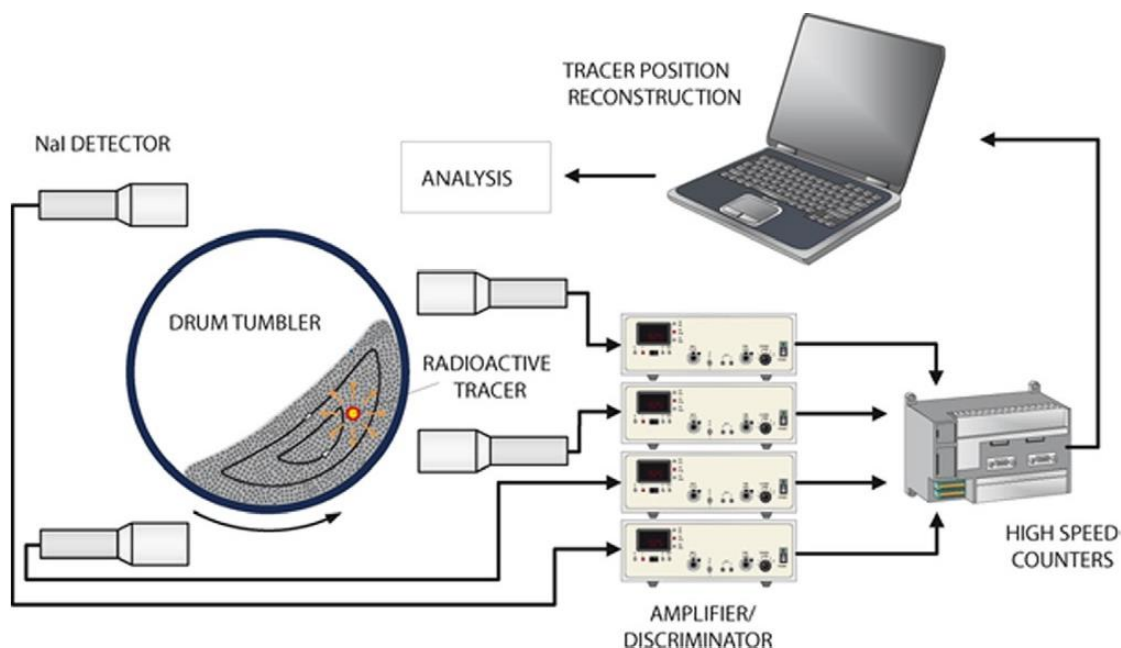
Figure 13: Typical RPT setup in drum tumbler. (Dubé et al. 2014)

The radiation detection and data acquisition system are expected not to be a limiting factor in the performance of radiotracer technique preprocessing systems, as a number of commercial systems are numerously available today.Ttechnological advancement in electronic parts and computing systems are combined with new architectures as an extensive development. Till date, this innovation is carried to allow high-performance systems that are suitable for the next generation of versatile radiation detection and data acquisition devices.

Conclusions

Radiotracer technologies clearly play a major role in process optimization, plant scaling up, design, and the performance of various chemical process industries by increasing the understanding of these processes and assisting in the modelling of their key multiphase flow systems. The industrial radioactive tracer techniques in general are invaluable tools for laboratory and pilot plant research.They are also useful for on-site industrial applications for diagnostics, monitoring, advanced mechanistic modelling development for performance prediction and optimization process, and for obtaining benchmark data for experimental evaluation and validation. Further investigation and development is needed to explore and strengthen these techniques, to develop new ones, and to improve the equipment and tools for the measurements and applications in industrial scale processes. In subsequent research, new development of industrial radioactive nanoparticle as the most appropriate tracer to be used in high temperature processes will be carried out because in practice there has been no stable candidate as a radiotracer over the temperature under pressure. The feasibility of industrial radioactive nanoparticles tracers will be estimated in terms of the efficiency in industrial applications.

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