Radiotracer method: A nuclear tool for flowrate measurement in the pipeline

S Sugiharto
Center for Isotopes and Radiation Application - Batan, Jl. Lebak Bulus Raya No. 49
Jakarta Selatan 12440, Indonesia
sugi@batan.go.id

Abstract. Experimental work has been carried out to demonstrate using the radiotracer method for flowrate measurement of water in a 3 inches diameter of the carbon-steel pipeline. The water was supplied from a water tank of a volume of approximately 5000 liters. The tank was connected to the pipeline through a flexible plastic hose. A ball valve was installed at the far end of the pipeline to adjust water discharge from the pipeline. None flow meters were installed on the pipeline. Iodine-131 (131I) radioisotope with a concentration of 1 mCi.mL⁻¹ has been injected instantaneously through the injection point located at 15 meters from the water tank. Two radiation detectors (notified as D1 and D2) which had been set-up respectively at the distance of 4 m and 7 m from the injection point recorded residence time distribution (RTD) curves of the radiation intensities emitted from the injected radioisotope. During the experiment, the system was assumed time-invariant by keeping the tank was always containing full of water. The transit time calculated based on mean residence time (MRT) was 89.9 s, and the linear velocity of the water flow is 3.33 cm.s⁻¹

1. Introduction
Fluid flow is commonly caused by the pressure difference at two or more different points of measurements. For industries and other strategic fluid installations, flow measurement is the most important physical quantity that has to be known for quantifying gases, liquids, and solids [1]. The most commonly used devices for flowrate measurement are termed flowmeters. The primary function of a flowmeter is to monitor, measure, or record fluid flow. The measurement modes are fluid velocities, pressure differences, or direct measurement of volume flow rate or mass flow. Many flowmeters are commonly used for flow measurements, and they are widely attributed by their name and sometimes under the same category. For instance, differential head meters of the orifice plate, flow nozzle, Ventry meter, flow tube, and Pitot tube is the meter types which are considered as linear flowmeters [2]. The essential thing in evaluating flowmeters is that they rely on the use of calibrated electronics. Therefore they are not globally standardized, and they shall be routinely calibrated at the predetermined time of servicing [2,3].

The two different approaches can describe fluid flow: by fluid mechanics theory [4] and by residence time distribution (RTD) one [5-8]. Meanwhile, fluid mechanics was known since the ancient Greek, and Roman times [4], the RTD concept was introduced later in the 1950s [5]. RTD is obtained from the injection of tracers into the inlet of the investigated system and records the injected tracer concentrations at its outlet. There are many kinds of tracers, but from a radioactivity point of view,
the tracers are divided into two main categories: non-radioactive tracers such as dyes, salts, conductive tracers, etc radioactive tracers or radiotracers for short. Radiotracer is a radioactive material which is used as tracers and produced in nuclear facilities [9, 10].

Although the RTD in today's perspective is considered an empirical yet straightforward approach to describe any processes involving fluid or particulate flow, RTD has been used as a useful tool for many purposes for troubleshooting and diagnosing the performance of industrial processing systems [11]. Scientists have reported the radiotracer method for flow rate measurement. They use radiotracer techniques for various purposes, among of which are used to measure the flow rate of the vapor phase or geothermal fluids in geothermal using Krypton-85 isotope [12], to determine the discharge rate of water in the open canal using Iodine-131 isotope [13], to determine the flow of water in a large diameter pipeline using Iodine-131 isotope [14], to determine flow rate of crude oil and water in enhancing oil recovery production using multi tracers method [15].

The advantages of using radiotracer for industrial and environmental applications are higher detection sensitivity that can be achieved because radiotracer uses high radiation energy of radioactive material. Therefore, detection can be carried out in the online mode because the high penetrating gamma energy of radiotracer can penetrate the enclosed wall of a system and many radiotracers for various flow applications in different phases [11,15]. Flow rate measurement is usually required for several following reasons: for calibrating the installed flow meters, for measuring flow rate in an industrial installation that does not have flow meters, and for measuring the flow distribution in a fluid installation network or for measuring pump's efficiency [16].

According to the International Organization for Standardization (ISO), the radiotracer method is best available for today's flow rate measurement techniques. It is better than conventional methods such as dyes and chemical tracers, which is generally very expensive. In many situations, water or gas flow measurement in a closed conduit using radiotracer can be executed using dilution or transit methods. The methods are based on measurement of the concentration-time distribution of radiotracer of known volume regardless of the massive flow rate of the liquid [17].

The purpose of this article is to introduce a method of flow rate measurement using the RTD concepts. A case study is given to demonstrate the measurement of water flow using Iodine-131 isotope as radiotracer in the small diameter of pipeline made of carbon steel materials. This study aims to introduce radiotracer techniques as a complementary method for flow rate measurement in industry using installed flowmeters.

2. Method

Mac Mullin and Weber first introduced the residence time distribution (RTD) concept in the analysis of chemical reactors [5]. Danckwerts then developed this concept further in his classical paper [18]. The work of Danckwerts formed the basis of various investigations involving flow system in chemical and biochemical reactors. Danckwerts introduced a fluid element that is an element of small volume concerning a system, but it is still large enough to accommodate sufficient molecules to define continuous properties such as density and concentration. His RTD theory is now using extensively by scientists for various research works and system investigation.

Residence time distribution (RTD) of fluid elements is defined as the spent time of such fluid elements in a system. The distribution of these times is called RTD function of the fluid, \( E(t) \), or \( E(t) \) curve which represents the fraction of fluid leaving the system at each time. \( E(t) \) is a fundamental indicator to describe how the extent of the flowing and mixing pattern in a chemical reactor is. It is also as an instrument for analysis of non-ideal reactors. The dimension of \( E \) is time\(^{-1}\). Mathematically, the RTD function is stated as

\[
E(t) = \int_0^\infty \frac{c(t)}{\overline{c}} \frac{d\tau}{c(t)}dt
\] (1)

where \( C(t) \) is the concentration of fluid elements at the outlet at a certain time \( t \).
The mean residence time (MRT), \( \tau \), is the first moment of the RTD distribution and formulated as [3,13]

\[
\tau = \int_0^\infty tE(t)\,dt \quad (2)
\]

Substitution Eq. (1) into Eq. (2) produces:

\[
\tau = \int_0^\infty \frac{tC(t)}{c(t)}\,dt \quad (3)
\]

For ideal system with constant flow rate, the mean residence time is equal to mean holding time for vessel [16]

\[
\tau = \frac{V}{Q} \quad (4)
\]

with \( Q \) is volumetric flowrate and \( V \) is the vessel volume.

In a continuous flow system, all fluid elements have their own residence time, therefore for sufficiently long period, all fluid elements will eventually come out from the system. This situation is called mass balance principle and is expressed as [6.11]

\[
\int_0^T E(t)\,dt = 1 \quad (5)
\]

By definition, Eq. (5) shows normalization of the residence time distribution function and fulfil the following condition:

- \( E(t) = 0 \) for \( t < 0 \) because there is no fluid entered or injected into the system.
- \( E(t) \geq 0 \) for \( t > 0 \) because fraction of fluid elements are always positive.

In general, an arbitrary distribution function is analyzed using statistical moments, but in many cases, the most commonly studied moment is the mean up to the fourth moment. The second central moment is variance as an indicator of the spread of the distribution. The third central moment is skewness and is used as an indicator of the distribution's asymmetry, and finally the fourth central moment or kurtosis is an indicator of how heavy-tailed the distribution is [19].

2.1. Selection of radiotracer

Selection of appropriate radioactive materials as a radiotracer for particular studies probably is the most important consideration because an isotope is not always suitable for all applications. The radiotracer's various characteristics include physicochemical behaviour, type and energy of radiation, the half-life of the isotope, specific activity, possibility of production, and radiotoxicity. Ideally, both radiotracer and traced material have the same physicochemical properties. When the traced material is the liquid phase, the selected appropriate radiotracer should be a liquid phase. In the majority of industrial applications, the injection of radiotracer does not involve chemical reaction with the traced materials, therefore the only physical properties of the radiotracer is needed to be considered. The required need is the ability of radiotracer mix thoroughly within the bulk material being traced. For online or 'in-situ' detection, the most suitable selected radiotracer is the gamma-ray emitting radiotracer. High energy of gamma radiation can penetrate the system's wall that makes it enable to be detected from outside of the wall. The half-life of the selected radiotracer should be considerably longer than the time needed for doing a complete experiment. Type and energy of radiation, radiotoxicity level and specific activity are related to the radiological safety point of view. The commonly used radiotracer for flowrate measurements are summarized in Table 1 [20].
Table 1. Commonly used radionuclides as radiotracer for flowrate measurement. [20]

| Radionuclide | Chemical form | Phase tracing | Half-life | Energies (MeV)(% ) |
|--------------|---------------|---------------|-----------|--------------------|
| $^{24}\text{Na}$ | Sodium Carbonat, Na$_2$CO$_3$(Na$^+$) | Aqueous water | | 1,369(100%); |
| | Sodium Nitrate, NaNO$_3$(Na$^+$) | Aqueous water | | 2,755(100%); |
| | Sodium Acetate, CH$_3$COONa | Organic phase | 15 h | 3,86(100%) |
| | Sodium Naphtenate, C$_{10}$H$_7$COONa | Organic phase | | |
| $^{41}\text{Ar}$ | Argon gas | Gas | 1.833 h | 1,294 (99%) |
| $^{82}\text{Br}$ | Ammonium bromide, NH$_4$Br(Br$^-$) | Aqueous water | 35.7 j h | 0,554(70%); |
| | Para-dibromobenzene, C$_6$H$_4$Br$_2$ | Organic | | 0,619(39%); |
| | Bromonaphthol, BrC$_{10}$H$_7$OH | Organic | | 0,698(25%); |
| | Bromodedecane, C$_{12}$H$_{25}$Br | Organic | | 0,776(85%); |
| $^{85}\text{Kr}$ | Kripton Gas | Gas | 10.6 y | 0,514 |
| $^{99}\text{Mo}$ | Sodium Molibdate (MoO$_4$$^-$) | Aqueous | 66 h | 0,14(88%); 0,74(14%); |
| $^{99m}\text{Tc}$ | Sodium Pertechnetate (TcO$_4$$^-$) | Aqueous | 6 h | 0,14(88%) |
| $^{131}\text{I}$ | Sodium Iodide, NaI(I$^-$) | Aqueous | 8.04 d | 0,364(80%); 0,64(9%); |
| | Potassium Iodide, KI(I$^-$) | | | |
| | dalam larutan sodium thiosulphate, Na$_2$S$_2$O$_3$ | | | |
| | sebagai agent pereduksi untuk iodine, | | | |
| | Iodo-benzene | | | |

2.2 Experiment
Radiotracer experiment for flowrate measurement was conducted in the laboratory of Nondestructive Investigation and Diagnostic at the Center for Isotopes and Radiation Application, National Nuclear Energy Agency of Indonesia (Batan). The experimental facility is a pipeline of 7.62 cm of diameter, which spans around 70 m length. One end of the pipeline is connected to a water tank of capacity of around 5000 liters. The water in the pipeline was supplied from a water tank that was always containing full of water. The volumic flow of water in the pipeline was regulated by adjusting the ball-type valve installed at the far end of the pipeline, making the water flow constantly due to gravity. The injection point was determined at the point located around 15 m from the water tank. The laboratory facility for radiotracer experiment is shown in Fig. 1. Iodine-131 isotope with a concentration around 1 mCi.mL$^{-1}$ was injected manually using plastic syringe into the pipeline. Two collimated scintillations NaI(Tl) detectors placed at distances 4 and 7 m respectively from injection point were used to record radiation intensity of injected isotope. Before the injection, these detectors were connected to datalogger and computers. There was no radiation detector placed close to the injection point because it would be affected by the isotope's gamma energy during transportation of the isotope from its container to the injection point and during the injection. The experimental data recorded by the NaI(Tl) scintillation detectors were residence time distribution (RTD) curves resulting from the pipeline's injected isotope. The recorded experimental RTD data were then be saved for further treatment and for data analysis. It worth noting that the dummy test was carried out in advance before performing the real experiment.
Figure 1. The laboratory facility for flowrate experiment using radiotracer method. I-131 isotope has been injected into the pipeline containing water flow supplied from water tank.

3. Result and discussion.
As mentioned before, the fluid flow can be learned from two approaches: fluid mechanic theory and residence time distribution (RTD) [5,18]. In fluid mechanics, fluid flow is usually formulated by the Navier-Stokes equation. Description of flow pattern in a continuous system is generally obtained from the Navier-Stokes equation solution theoretically. However, it is not easy to get a satisfactory answer because the flow is too complicated, even in a single phase. In a microscale point of view, fluid flow in axial direction will generate radial flow due to mass and momentum transfers among the fluid molecules. Moreover, these molecules’ interaction and boundaries may generate eddies that embody a turbulent regime [22]. In such a situation, a numerical solution based on the stochastic process is recommended to obtain the solution.

RTD which based on statistical theory, gives an alternative solution for flowrate learning. It is found by injecting radiotracer at the inlet and monitoring the tracer concentration at outlet of the system being investigated. In early development, two ideal flow models were commonly used to solve fluid flow problems, namely plug flow (PF) and continuous stirred tank (CST). However performance of fluid flow in real system is in between these two ideal models. The fluid flow in real system were first approached by Danckwerts based on RTD analysis [18].

To implement RTD method in a continuous flow system, Danckwerts and Nuauman [5] introduced several primary assumptions from which both experimental and mathematical understanding can be derived. These assumptions are fundamental to characterize fluid flow, these assumptions are: (1) the system being studied must continuous. Thus removal fluid component to and from the system does not influence its continuity properties; (2) the material within the system is homogeneous when the materials is multiphase, it must be immiscible; (3) the flow at the inlet and outlet of the continuous flow system is fully developed and steady-state which indicate that the system is invariant throughput time or repeatable period of time; (4) the bulk flow at inlet and outlet of the system have a unidirectional flow, so that once material and radiotracer enter the system they reside within until leaving the system and never to return; (5) the system is isothermal; and (6) addition of the radiotracer material into the system does not affect the system's overall flow and the radiotracer is eventually distributed along the entire system's cross-section. This assumption is very important to minimize error.
In this study, the assumptions mentioned above are applied for both tracer selections and system characteristic. Moreover, the following radiotracer properties concerning the bulk flow are required: (i) the radiotracer must be miscible and have physical properties similar to those of the fluid under investigation, (ii) the radiotracer should be accurately detectable in small amount and introduction of radiotracer into the system do not affect the flow of traced fluids, (iii) the radiotracer concentration should easily be monitored and the recorded radiation intensity should be proportional to its concentration, and (iv) sorption of the injected radiotracer by the internal wall of the system should not be occurred. In the radiotracer experiment, the use of highly sensitive detector is required for capable of detecting very low limit radiotracer concentration and the accurate detection is highly dependent on tracer type and concentration. [21]

3.1 Data treatment

The experimental data which has been transformed into the excel datasheet, is presented in Figure 2. Several treatments to the raw experimental data were carried out. The radiation detectors record all radiation that comes to them, including background radiation and the radiation originates from the injected isotope. The background radiation is random process and is usually considered as noise. Its contribution is to give additional recorded radiation by the detector but do not give valuable information to the whole recorded data. In this experiment, the background radiation was eliminated and it provides a base-line of the RTD curves.

A radiotracer is a radioactive material that is decayed due to its radioactivity properties. The radioactivity's representative quantity is half-life, representing the time required for radiotracer to reduce to a half of its initial radioactivity. As the half-life of Iodine-131 isotope is around 8 days, whereas the time required for doing a complete experiment is only less than 1 hour, therefore the data correction due to radioactivity is not necessarily be carried out. Long tail of RTD curve may indicate a sticky zone or dead zone in the pipeline wall on which the radiotracer resides there temporarily longer than those of the main flow.

3.2 Flowrate measurement

The experimental work's objective is to determine the flowrate of water in a pipeline of small diameter. There are two methods for determining flow velocity from analysis of RTD curves, namely the peak to peak method and by calculating RTD mean residence time (MRT). In the peak to peak method the 'peak' of the RTD curves at each position of the detectors are determined and the time required by the radiotracer to move from one detector to the other detector is calculated [3]. If $t_1$ and $t_2$ are respectively the peak times of the RTD curves at the first and the second detector position, and $L$ is the distance between the two detectors, the flowrate, $v$ is calculated by the following equation [3]

$$v = \frac{L}{t_2 - t_1}$$

Determination of flowrate using peak to peak method can give adequate calculation provided that the RTC curve is smooth, relatively slim and has one peak. The generated RTD from the radiotracer experiment in a real system is usually not smooth instead fluctuate due to flow dynamics in the system and the injected radiotracer's radioactivity properties. The flowrate calculation may probably be better established using MRT method because it represents the curve's gravity center [22]. The MRT as expressed in Eq. (3) is the integral form which need to be re-expressed numerically as

$$\tau = \frac{\sum_{i=1}^{n} t_i c(t)}{\sum_{i=1}^{n} c(t)}$$

The denominator of Eq. (3) represents area under curve and is calculated using integral calculus method one of which is using the Simpson's method [23]
In this experiment, 900 data was recorded with time base measurement was set for each 1 second. The measured data at each detector position shown in Fig. 2 consists of radiation intensities from background radiation and injected radiotracer. Truncation to the RTD data was executed. The evaluated RTD data are those of the RTD curves originate from the curve's rise until its return to background levels. Based on numerical data, as shown in Fig. 2 there were 299 and 432 evaluated data in RTD D1 and RTD D2, respectively. The mean residence time (MRT) was calculated using Eq. (3) based on the assessed data. The flowrate was calculated using Eq. (6), with the denominator is the transit time of the radiotracer moving from one detector to the other. The transit time calculated based on calculated MTR was 89.9 s. As the distance between the installed sensors was 3 m, the flowrate of the linear velocity of the water flow in the pipeline is around 3.33 cm.s$^{-1}$, as the system being studied is considered time-invariant. The physical quantities calculated in this work is summarized in Table 2. As mentioned elsewhere [22], the shape and spread RTD data give insight into the system's flow phenomena. Here, spread data on RTD D2 is larger than RTD D1, indicating that some mixing process occurred between the water and the injected radiotracer.

![Figure 2. RTD experimental data obtained from injection of I-131 isotope for flowrate measurement.](image)

| No | RTD  | Analysed data | Number of analysed data | MRT (s) | Flowrate (cm.s$^{-1}$) |
|----|------|---------------|-------------------------|---------|------------------------|
| 1  | RTD D1 | 334 to 633    | 299                     | 100.8   | 3.33                   |
| 2  | RTD D2 | 414 to 846    | 432                     | 110.7   |                        |

4. Conclusion
A laboratory radiotracer experiment has been demonstrated to measure water flow in a small diameter pipeline using Iodine-131 isotope. The calculation of physical quantities was based on the assumption that the studied system was time-invariant and linear. The calculated linear velocity of water flow or flowrate was 3.33 cm.s$^{-1}$. The mixing process between water flow and injected radiotracer occurred as indicated by the spreading of RTD curves.

Acknowledgement
The Government of Indonesia funded the research work under the government's research funding 2018 for the Center for Isotopes and Radiation Applications for.

References
[1] Liptak B G 2003 *Instrument Engineers' Handbook, Vol 1: Process Measurement and Analysis* (Boca Raton: CRC Press) ep 2
[2] Cran Co 2013 *Flow of fluids through valves, fitting and pipe* (Connecticut: Technical Paper No 410) cp 4

[3] Kasban H Ali E H and Arafa H 2017 *Nuclear Engineering and Technology* **49** 196

[4] Escudier M 2017 *Introduction to Engineering Fluid Mechanics* (Oxford: Oxford University Press)

[5] Nauman E B 2008 *Ind. Eng. Chem. Res.* **47** 3752

[6] Fogler H S 2004 *Elements of Chemical Reaction Engineering* (New Delhi: Prentice-Hall of India)

[7] Pant H J and Sharma V K 2015 *Appl. Radiat. Isotopes* **99** 146

[8] Pant H J Sharma V K Shenoy K T and Sreenivas T 2015 *Appl. Radiat. Isotopes* **97** 40

[9] Lee S Chung S and Park S 2007 *Journal of Nuclear Science and Technology* **44** 1467

[10] Kasban H and Hemed A 2014 *J. Radioanal. Nucl. Chem* **300** 379

[11] IAEA 2008 *Radiotracer residence time distribution method for industrial and environmental application, Training Course Material*, 31 (Vienna: IAEA)

[12] Sugiharto S Wibisono W Kushartono K Achdiyat A Azni B Suryantoro T Y Ani A and Abidin Z 2014 *Atom Indonesia* **40** 89

[13] Pant H J Goswami S Biswal J Samantray J S and Sharma V K 2016 *Applied Radiation and Isotopes* **112** 89

[14] Biswal J Pant H J Goswami S Samantray J A Sharma V K and Sharma K S S 2018 *Flow Measurement and Instrumentation* **59** 194

[15] Sugiharto S Su‘ud Z Kurniadi R Wibisono W and Abidin Z 2009 *Applied Radiation and Isotopes* **67** 1445

[16] Pant H J Biswal J Goswami S Samantray J S Sharma V K Sharma K S S and Sukla S 2017 *BARC Newsletter* pp.1-9

[17] ISO 2975-1 1974 *Measurement of water flow in closed conduits-Tracer methods-Part 1:General*

[18] Danckwert P V 1953 *Chem. Eng. Sci.* **2** 1

[19] Taylor J K and Cihon C 2004 *Statistical Techniques for Data Analysis*, 2nd edition (Boca Raton: Chapman & Hall/CRC)

[20] IAEA 2004 *Report of a consultative meeting held in Warsaw, Poland, 16-19 June*

[21] Torres A P and Oliveira F A R 1998 *Journal of Food Engineering* **36** 1

[22] Sugiharto S Stegowski Z Furman L Suud Z Kurniadi R Waris A and Abidin Z 2013 *Computer and Fluids* **79** 77

[23] Larson R and Edwards B H 2010 *Calculus*, 9th edition (USA: Brooks/Cole) p 314