MINING AND ENVIRONMENTAL PROTECTION

PROCEEDINGS

Editor
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25 - 28. September 2019.

FOREWORD

After the consultations with business entities in the field of mining and environmental protection, faculties and scientific institutes, an initiative for organizing a scientific meeting on mining and environmental protection was taken in 1996. The Faculty of Mining and Geology in Belgrade, CENTER FOR ENVIRONMENTAL ENGINEERING, have organized the First Yugoslav Conference with International participants held from 25th to 27th April 1996 in Belgrade, Serbia. 2nd International Symposium was held in Belgrade from 25th to 27th 1998. 3th Symposium was held in Vrdnik from 21st to 23rd May 2001, 4th International Symposium was held in Vrdnik from 23rd to 25th June 2003, 5th International Symposium was held in Vrdnik from 10 to 13 June 2015 and 6th International Symposium was held in Vrdnik from 21st to 24th June 2017.

Due to the large number of subjective and objective reasons organization of the symposium was discontinued in 2003. On the basis of the conclusions made at the 6th Symposium MEP 2017 and great interest of domestic and foreign scientific and professional public, the Faculty of Mining and Geology in Belgrade, in cooperation with co-organizers (National University of Science and Technology “MISIS”, Moscow, Russia Berg Faculty TU Košice, Slovakia, University of Ljubljana, Faculty of Natural Sciences and Engineering, Slovenia, Goce Delčev University in Štip, N. Macedonia and University in Banja Luka, Faculty of Mining, Prijedor, Republic of Srpska, Bosnia & Herzegovina), is organized the 7th International Symposium Mining and Environmental Protection – MEP 2019.

The previous Symposium were very successful and scientist and companies from many countries gathered to exchange information and research results. The objective of this Conference is to bring together engineers, scientists and managers working in mining industry, research organizations and government organizations, on development and application of best practice in mining industry in the respect of environment protection.

At the Book of Proceedings of 7th International Symposium on Mining and Environmental Protection are 56 Papers. Almost half is from abroad, or their authors is from different countries. At least 150 authors and co-authors took part in the preparation of these papers. The papers were reviewed by Reviewers and Scientific Committee. Only high-quality papers were selected, from two side, one from the scientific basis and the second from point of view of applicability in resolving problems at the development of mining.

We are very grateful to the authors of the papers, who contributed to a great extent to the success of this meeting by having sent enough number of high quality papers, and thereby made the work of the reviewers a pleasant one in respect of selecting the best quality papers. Also, we would like to thank all participants in the Symposium, as well as the sponsors who helped and enabled us to hold such a great meeting.

Editor

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TECHNETIUM-99M AS A TRACER FOR THE LIQUID RESIDENCE TIME DISTRIBUTION MEASUREMENT: OPTIMIZATION OF DIFFERENT RADIOTRACER PARAMETERS FOR FLOW METER CALIBRATION

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Abstract: *Flow meters are essential part of equipment in almost all chemical and process industries. Knowledge of the flow dynamics and flow of material through the reactor system is required to evaluate the performance of the reactors, processes, operation and design. Traditionally this evaluation can be performed using chemical tracers. Chemical tracer results are not always precise and they could not allow an online diagnosis. The samples containing chemical tracer have to go to a lab for analysis and chemical tracers are less sensitive than radioactive ones. Radioactive tracers are the only tracers capable of measuring the active residence time distribution (RTD) with high degree of precision and give information on-line without stopping the process. In this work the application of radiotracer method for RTD measurement in flow meter calibration is described. The RTD measurements of the system were performed in pilot-scale perfect mixer in series reactor for flow meter calibration with Technetium-99 m (^{99m}Tc) as a radiotracer in the form of pertechnetate ion ($^{99m}TcO_4^-$). The optimization of the parameters included input signal of radiotracer, concentration of radiotracer and position of detectors. The measured data were analyzed, and standard and relative deviations were quantified under different operating conditions. A plug flow model was used to simulate the measured RTD curves and investigate the flow dynamics of the flowing water. The results of the study showed that the optimum concentration of ^{99m}Tc for flow meter calibration was 0.3 ml having activity 0.68 mCi. The standard deviation was σ=0.10 l min^{-1} and the relative deviation was 0.94%. The results of the study can help in the operation and design of the existing systems and design of new ones.*

Keywords: *Radiotracers, RTD, Technetium-99m, Flow meter*

1. INTRODUCTION

In chemical engineering, an equipment is designed to behave as either an ideal plug flow reactor (PFR) or an ideal continuously stirred tank reactor (CSTR) depending upon the process requirement. However, in actual operating systems, the flow patterns deviate from the designed ideal flow patterns because of the occurrence of one or more abnormalities such as bypassing, dead zones and channeling [1]. These abnormalities cause deviation in the flow patterns and eventually lead to deterioration in product quality and decrease in process efficiency, thereby causing significant economic loss for the industry and even posing a threat to the manufacturing plants and the environment [2,3]. Measurement and analysis of residence time distribution (RTD) of process material is an important approach to identify abnormalities, if any, and to investigate the hydrodynamics of process equipment in the chemical process industry. The approach is very reliable and provides valuable information about the performance of process equipment such as reactors and columns [4]. This is also used for the evaluation and optimization of the design of laboratory and pilot-scale systems at the design stage. This approach is also used in evaluating and calibrating equipment, whether it is a reactor, pipe or flow meter.

Conventional tracer techniques using dyes, chemicals and salts are often used to measure the RTD of process fluid in laboratory-scale systems; however, they cannot be applied to pilot-scale and large-scale industrial systems because of their many disadvantages [5-7]. The disadvantages of the conventional tracer techniques are overcome by radiotracer by their high detection sensitivity, physico-chemical compatibility, online detection ability and availability for most of the phases in industry [8-10]. In addition to this, multiple radiotracers can be simultaneously used for tracing different phases because emission of characteristic gamma radiations can be detected by spectroscopic measurements [11].

It is known that the RTD measurement approach using radiotracers has immense potential and thus has been used extensively to characterize flow in different industrial systems [2, 12-20]. However, data used in process calculations usually need to be verified (volume of the reactor, flow of the stream etc.) in order to perform accurate measurements and calculations. Flow meters are essential part of equipment in almost all chemical and process industries. Knowledge of the flow dynamics and flow of material through the reactor system is required to evaluate the performance of the reactors, processes, operation and design.

In this work the application of radiotracer method for RTD measurement in flow meter calibration is described. The RTD measurements of the system were performed in pilot-scale perfect mixer in series reactor for flow meter calibration. Technetium-99 m (^{99m}Tc) in the form of pertechnetate ion ($^{99m}TcO_4^-$) was used as radiotracer.

2. EXPERIMENTAL

The application method of the radiotracer experiment to flow meter calibration is shown in Figure 1. The principle of radiotracer experiment consists of injection of an appropriate quantity of a specific radiotracer at the inlet of the pipe and recording, with gamma ray detector, the count rate (counts/s)–time curve at the outlet. Detectors should be placed at minimum 50 diameters of pipe away from the inlet, but it should be preferably 100 diameters of pipe. The radiotracer photon count rate versus time at the outlet of the system after normalization is the experimental RTD.

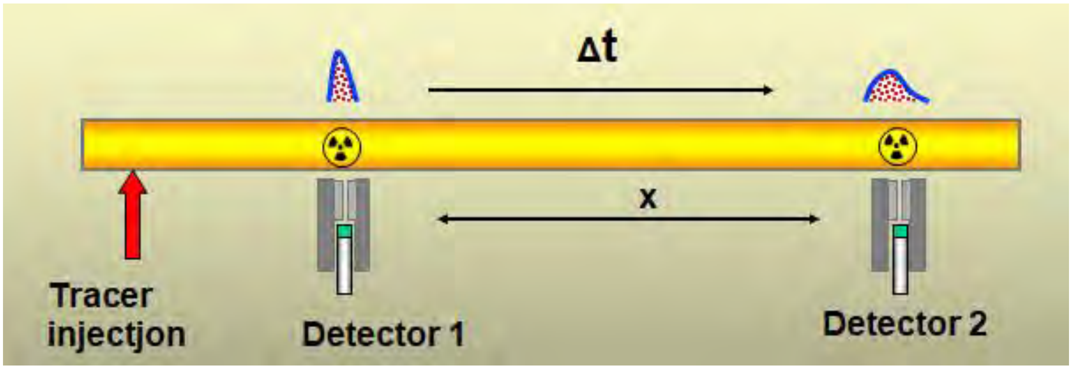


Figure 1. Schematic layout of radiotracer experiment applied to flow meter calibration experiment: injection of a radiotracer (which emits gamma rays) at the inlet of the pipe and recording (using a gamma ray detector) the count rate (counts/s)–time curve at the outlet. The obtained function is the experimental RTD.

Schematics of the laboratory scale flow rig for flow meter calibration and flowrate measurements is presented in Figure 2. Few possible placements of detectors (probes) are shown in the Figure 2.

For the RTD measurements and flow meter calibration, water at 22°C as working fluid was used.

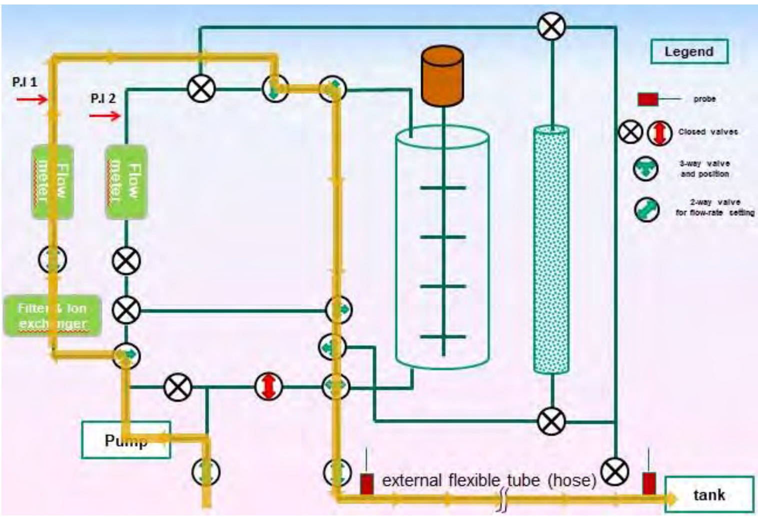


Figure 2. Flow rig setup for flow rate measurements and flow meter calibration.

Several characteristic parameters of the reaction process could be obtained directly from the experimental RTD such as mean residence time (s), flow rate and mixing intensity. We are interested in flow rate. RTD could be a precious source of information about reactor troubleshooting—parallel flows, stagnant zones or bypasses. However, Thyn and Zithny [21] noted clearly in their work that the applications of radiotracers for determination of RTD as well as technical conditions for realization of experiment in industry create special requests for data processing. Therefore many conditions should be considered to make a success such experimental method of on-line diagnostic.

2.1. Optimum radiotracer

Over 3,000 radionuclide varieties are listed in published compilations [22, 23]. Practically, about 100 species are commonly used as radiotracers. A number of essential requirements have to be balanced in choosing the optimum radiotracer:

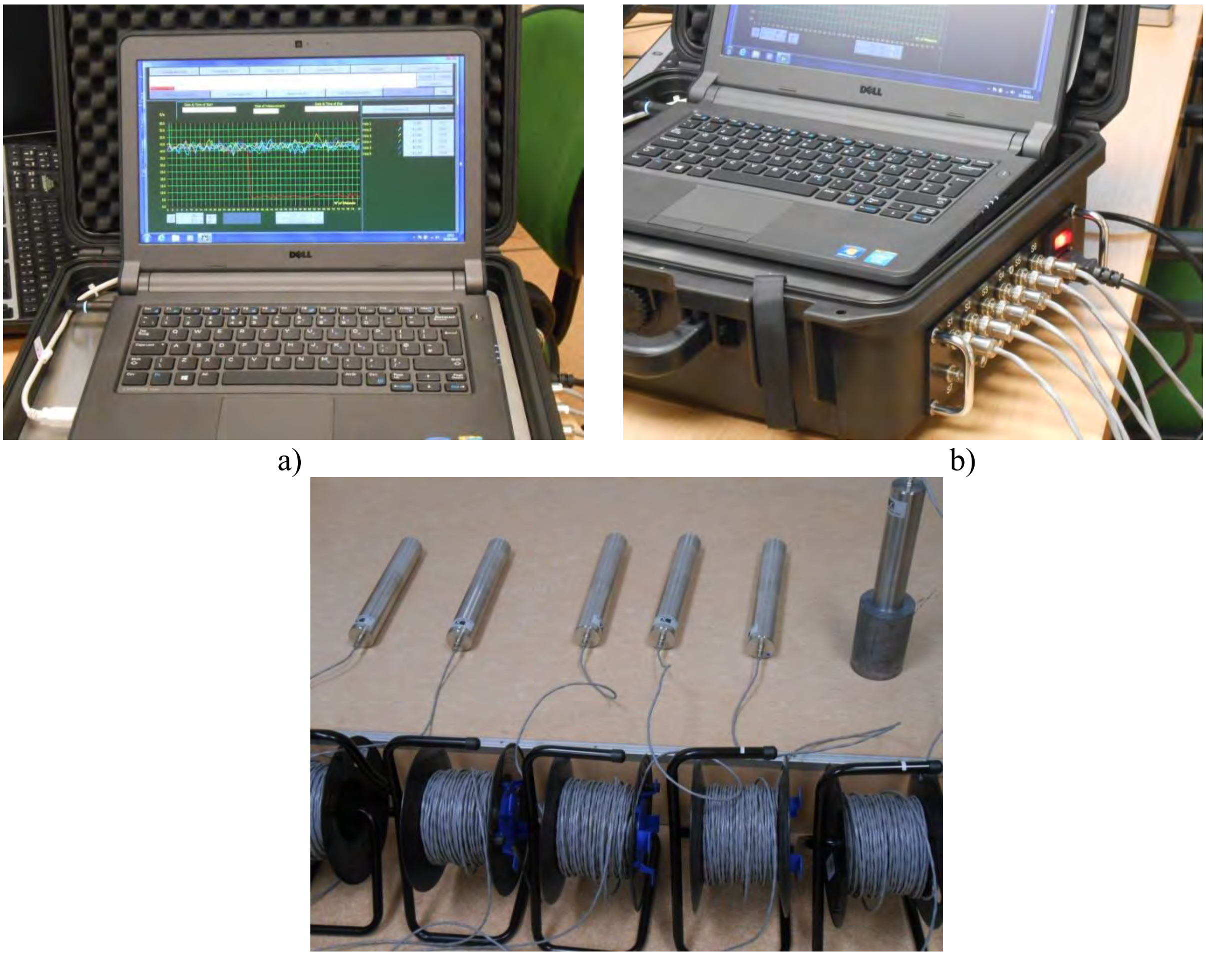
- The integrity of the radiotracer: the radiotracer should behave in a similar way the loading behaves in the process reactor which is under study. The physical and chemical properties of the radiotracer should be well examined. Especially essential are the boiling and evaporation temperatures, the complete miscibility of the radiotracer with the substance to be traced as well as its resistance against thermal decomposition and its chemical stability under the conditions of the investigated process. If it is not the case the radiotracer behavior will not be representative of flow in the reactor. Hence, and taking into consideration the work of Gallorini et al. [24] on how to check the specific characteristics, stabilities and purities of the radiotracers, the suitability of the chosen radiotracer must be confirmed by validation tests in the laboratory.
- The nuclear properties of the radiotracer: to get efficient detection, the radiotracer should emit a gamma ray with adequate energy and yield. A radiotracer with the shortest half-life should be chosen to minimize radiological impact whilst ensuring that detectable life of the radiotracer is sufficient to match the timescale of the investigated reaction process being studied.
- Radiological and environmental aspects: the as low as reasonably achievable (ALARA) [25] principle should be respected during the preparation and handling of the radiotracer to prevent radiological impact on operators. Schonhofer et al. [26] noted that radiation protection considerations should not be forgotten and any unnecessary impact has to be avoided. Hence even if the radiotracer seems the best one for detection efficiency, in fact it will be the worst if it will involve environmental impacts that are not adequately reflected in human assessments.

A meticulous work of research for the optimum radiotracer was applied by Colbert et al. [27] to characterize the behavior of hazardous elements during waste treatment. For investigation of flow meter calibration experimental tests with ^{99m}Tc was performed. $_{99m}$Tc is eluted from ^{99}Mo/^{99m}Tc medical generator. It emits 140 keV gamma rays with an efficiency of 89%. The exposition dose from ^{99m}Tc with a radioactivity of 27 mCi is small in comparison with other radiotracers. The half-life of ^{99m}Tc (6.0 h) was more than sufficient to allow for complete evaluation and calibration of flow meter in performed tests. The short half-life and the rapid dispersion of ^{99m}Tc combine to ensure that there are no adverse effects on either the operator or the environment.

2.2. Detection system

A number of NaI(Tl) detectors of gamma radiation should be well fixed and collimated at the outlet of the process under diagnose. Detector's collimation is an essential condition to get a correct and readable RTD curve. Detector should have a small acceptation angle through which count rate is recorded (see Figure 3c). Also it is highly recommended to avoid detector's placements near outlet pumps; such placements are a source of trouble in the RTD measurement: the detector's signal will have a periodic disturbance depending on the pipeline flux imposed by the nearest pump function. Once all needed detectors are fixed, collimated and cabled to the laptop of the data acquisition system (DAS), and prior to radiotracer preparation and injection, an acquisition of the background radiation level is necessary to estimate through detector's signals the amount of activity detected. For a high accuracy, a maximum count rate should be about 100 times larger than the background radiation counts. Therefore a minimal background signal is required for each detector.

The detectors with cabling, as well as DAS with background radiation data acquisition are shown in Figure 3.

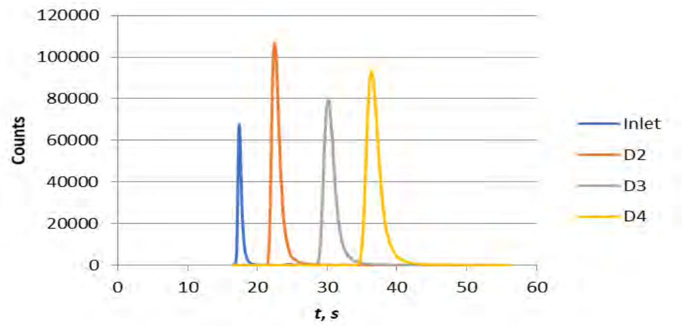


a) b) c)

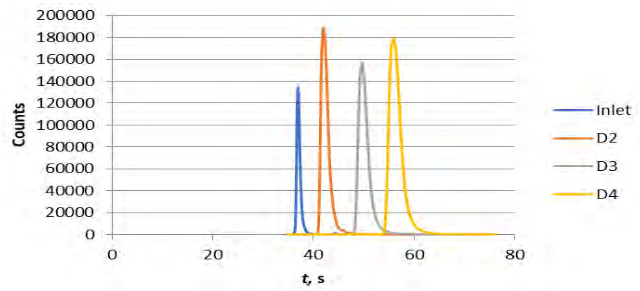
Figure 3. a) Front view of the data acquisition system (DAS) with laptop and data acquisition and treatment software CAESAR 12, b) 6 detectors are connected to DAS, c) 6 NaI(Tl)-detectors with cable drums (each drum contains about 50 m wire for signal transfer to the data acquisition system). One of the detectors is positioned in a lead collimator

3. RESULTS AND DISCUSSION

Figure 4 presents some of the experimental result of the tests that were performed on the lab scale equipment for flow rate measurements and for optimizing parameters for calibration of flow meters. Figure 2a is for 0.3 ml of ^{99m}Tc having activity 0.68 mCi, while Figure 2b is for 1 ml of ^{99m}Tc having activity 2.25 mCi. The count rate (counts/s)–time curve at the outlet was measured at four collimated NaI(Tl) detectors (D1-D4) positioned at different positions, D1 at the radiotracers inlet and D2, D3 and D4 at outlet. Each of the measurements performed at flow rig had the same instantaneous impulse (Dirac) input of radiotracer in the system, which can be seen in Figure 4.



a)



b)

Figure 4. RTD curves recorded by detectors placed at radiotracers inlet and at 50, 70 and 100 diameters of pipe length for 99mTc volume of a) 0.3 ml having activity of 0.68 mCi and b) 1 ml having activity of 2.25 mCi

The data treatment of RTD curve should begin by subtracting background level. Then a mathematical interpolation method should be adopted to correct disturbed sections of the RTD curve. Also a mathematical extrapolation of the RTD curve is done. Once the previous corrections (background subtraction, interpolation, extrapolation) are done to the experimental RTD curve, statistically, the RTD function is defined such that E(t)dt is the fraction of the total radiotracer amount that reside in the reactor for a time between t and t-dt. If C(t) represents the radiotracer count rate (cps) measured at the time t in the outlet of the diagnosed reactor, then RTD function is defined as follows:

$$E(t) = \frac{C(t)}{\int_0^\infty C(t)\mathrm{d}t}$$

For flow rate measurements axial dispersed plug flow was used, as the best fitting for the data:

E(t)

$$= \frac{\sqrt{\tau P_e}}{2\sqrt{\pi t^3}} e^{-P_e(\tau - t)^2/4\tau \times t}$$

where τ is the mean residence time, Pe is the Peclet dimensionless number and t is the time dimension.

Figure 5 shows the RTD software solutions to the experimental data for axial dispersed plug flow for ^{99m}Tc volume of 0.3 ml having activity of 0.68 mCi measurements.

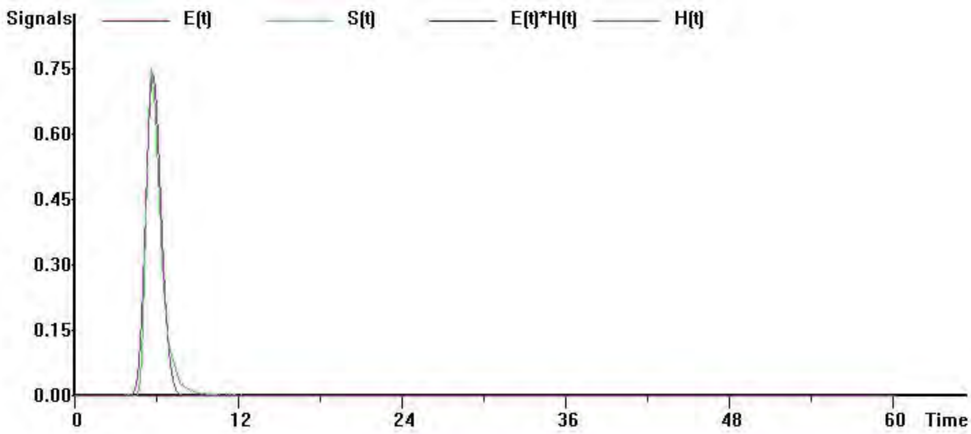


Figure 5. RTD software solutions to the experimental data for axial dispersed plug flow for 99mTc volume of 0.3 ml having activity of 0.68 mCi measurements. S(t) represent measurement, E(t) is model curve and H(t) represent the error.

Tests were performed for volume of ^{99m}Tc of 0.1 ml, 0.3 ml, 0.5 ml, 0.7 ml and 1.0 ml having activities of 0.22 mCi, 0.68 mCi, 1.12 mCi, 1.58 mCi and 2.25 mCi. Calculated flow rates, calculated from 3 independent measurements and 3 different detectors that can be used for flow meter calibration are presented in Table 1. Table 1 shows average flow rate and standard deviation of the results.

Table 1. Flow rates with standard deviation for different 99mTc volumes

V [mL]	u_{exp}[l min^{-1}]	σ_{exp}[l min^{-1}]
0.100	11.09	0.13
0.300	11.13	0.10
0.500	11.04	0.12
0.700	11.07	0.14
1.000	11.11	0.14

Representation of relative deviation is presented in Figure 6.

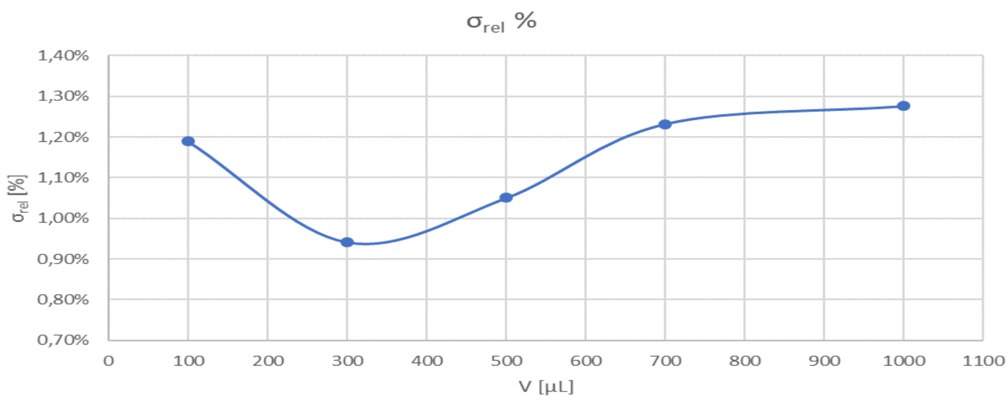


Figure 6. Relative deviations for different volumes of 99mTc in flow meter calibration tests

It can be seen from Figure 6 that the optimal condition for calibration of flow meter, giving the smallest standard deviation and relative error is for volume of ^{99m}Tc of 0.3 ml having activity of 0.68 mCi.

4. CONCLUSIONS

The RTD measurements of the system were performed in pilot-scale perfect mixer in series reactor for flow meter calibration with Technetium-99 m (^{99m}Tc) as a radiotracer in the form of pertechnetate ion ($^{99m}TcO_4^-$). The optimization of the parameters included input signal of radiotracer, concentration of radiotracer and position of detectors. The measured data were analyzed, and standard and relative deviations were quantified under different operating conditions. A plug flow was used to simulate the measured RTD curves and investigate the flow dynamics of the flowing water. The results of the study showed that the optimum concentration of ^{99m}Tc for flow meter calibration was 0.3 ml having activity 0.68 mCi. The standard deviation was σ=0.10 l min^{-1} and the relative deviation was 0.94%. The results of the study can help in the operation and design of the existing systems and design of new ones.

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