Optimization of injected radiotracer volume for flow rate measurement in closed conduits

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Abstract
In chemical processes it is essential that the flow in the process is accurately defined. Fluid velocity measurements are important for fluid flow quality performance in flow systems. This study focuses on determination of the volumetric flow rate and its standard (relative) deviation for calibration of conventional flow meters by using a radiotracer approach. The measurements for flow meter calibration were performed at a pilot-scale flow rig using Technetium-99m (99mTc) as a radiotracer in the form of pertechnetate ion (99mTcO4-). The measured data were analyzed, and precision of the experimental setup was investigated under two different approaches – IAEA’s RTD software and sum approximation of raw data. For the first time, the variation of standard deviation of calculated flow rate with the injection volume and activity of the radiotracer was determined. Plug flow with axial dispersion was used to simulate the measured RTD curves and investigate the flow dynamics of the flowing water. The results of the study have shown the possibility of in situ calibration of flow meters with a relative error lower than 1%. They also revealed a slight dependency of the precision of output results on the injection volume as well as similar results for manual and specialized RTD software data processing.

Keywords: radiotracers; RTD; technetium-99m; flow meter; calibration; chemical industry.

1. INTRODUCTION
Flow meters are essential part of equipment in almost all chemical and process industries. Knowledge of flow dynamics of materials passing through a reactor system is required to evaluate the performance of single reactors or even the whole processes. Traditionally this evaluation can be performed by using stable tracers. Results obtained in chemical tracer studies are not always precise and such studies cannot provide online diagnosis. Samples containing the chemical tracer usually need to be analyzed off-site, which requires additional time and logistics. In addition, stable tracers are in general less sensitive than radioactive ones. Radioactive tracers are suitable for measuring the active residence time distribution (RTD) with a high degree of precision and for on-line analysis that does not affect the continuity of the process.

It is well known that reaction equipment is designed to behave either as ideal plug flow reactor (PFR) or ideal continuously stirred tank reactor (CSTR). Choice of the reactor type depends on the process requirements. However, it is well known that flow patterns observed in reactors in practice deviate from ideal flow patterns because of anomalies such as bypassing, dead zones and channeling of the reaction fluids [1]. These deviations in flow patterns lead to deterioration in product quality and decrease in process efficiency resulting in significant economic losses for industry and eventually threatening their survival and environmental safety [2,3]. Therefore, an important approach to identify
flow anomalies and to investigate hydrodynamics of the process equipment in the chemical process industry is to measure and analyze RTD in the process. This approach is very reliable providing valuable information about the performance of process equipment such as reactors and columns [4]. Also, data obtained from the RTD measurement are useful for the design, evaluation and optimization of laboratory- and pilot-scale systems.

Conventional tracer techniques using dyes or other chemicals are often used to measure the RTD of fluids in laboratory-scale systems. They cannot be applied to pilot-scale and large-scale industrial systems because of their many disadvantages [5–7], such as low detection sensitivity, required physical-chemical compatibility and unsuitability for online detection for most of the phases in industrial processes. These disadvantages of the conventional tracer techniques are overcome by radiotracers [8–10]. Furthermore, multiple radiotracers can be simultaneously used for tracing different phases because emission of characteristic gamma radiations can be detected by spectroscopic measurements [11].

The RTD measurement approach using radiotracers has immense potentials and thus has been used extensively to characterize flow in different industrial systems [2,12–29]. However, data used in process calculations usually need to be precisely determined (volume of the reactor, flow rate of the stream, etc.) in order to perform accurate calculations. Independent measurement of the volumetric flow rate is known to be the desirable correction method to calibrate installed flow meters [20,21]. Flow meters are an 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 reactors, whole processes and their design.

Reactor flow anomalies, which include parallel flows, stagnant zones or bypasses, can easily be detected from the analysis of RTD data. However, it should be noted that special requests for data processing must be met for successful online diagnostics [22,23]. Several characteristic parameters of the reaction process could be obtained directly from the experimental RTD, e.g. the mean residence time (MRT) or mixing intensity. Therefore, flow rate, as a function of MRT and the volume of the system, can be easily calculated from such measurements.

In this work, the application of the radiotracer method for RTD measurement was used to calibrate a flow meter within a pilot-scale water flow-rig. The calibration experiments were carried out using different volumes (activities) of radioactive pertechnetate $^{99m}$TcO$_4^-$ while standard and relative deviations of calculated flow rate were investigated for both calculation methods.

2. EXPERIMENTAL

2.1. Experimental set-up

Schematics of the laboratory scale flow-rig used for the flow meter calibration and flow rate measurements is presented in Figure 1. The peak to peak method (PPM), which was employed in the experiment, involved the injection of a suitable radiotracer into the process stream to measure the volumetric flow rate of liquid in the system. To date, the peak to peak method is preferable and feasible for RTD determination in pipes with the constant internal diameter.

The volumetric flow rate, $Q$, is then defined as:

$$Q = \frac{Ad}{\Delta t} = \frac{\pi r^2 d}{\Delta t}$$  \hspace{1cm} (1)

where $A$ is the cross-section area of the pipeline with the radius $r$, $d$ is the distance between the two detectors mounted on the pipe line and $\Delta t$ is the transit time of the tracer between the two detectors.

The depicted radiotracer experiment (Fig. 1) consisted of injection of the appropriate quantity of $^{99m}$TcO$_4^-$ solution at the injection point 1 (IN 1), followed by immediate gamma detection via NaI(Tl) detectors positioned both right after the injection point (D1) and before the outlet (D2–D4). The detectors were mounted externally on the pipes and collimated using a lead sheet to eliminate surrounding background radiations. In order to allow the tracer to be homogenized over the whole pipe cross section, the injection point should be positioned at the distance equal to nearly 100 times of the diameter before the first detection point, yielding nearly 100 cm in the laboratory setup (pipe diameter of 1 cm) [11]. Thus, three detection probes at the outlet were positioned at lengths equal to 50, 70 and 100 times the
pipe diameter. In fact, the first two detectors are sufficient for the flow rate measurement using the transit time method, the third detector was placed for comparative and better statistics of the value. The radiotracer gamma count rate vs. time at the outlets of the system presenting the experimental RTDs, i.e. three RTDs (responses of D2, D3 and D4) were produced in one run resulting in three MRT values. Data from D1 were used as the injection-quality control and as an input for RTD analysis.

2. Detection system

Four NaI(Tl) gamma probes (Crytur, spol. s r.o., Czech Republic); at D1 after the injection point and 3 probes at the outputs D2-D4) were well fixed and collimated along the pipe length of interest for more data outputs in one run. Detector collimation is an essential condition to get a correct and readable RTD curve because it provides a small acceptance angle through which the count rate is recorded, i.e. not generating false signals obtained from the surrounding radioactivity. Also, it is highly recommended to avoid detector’s placements near pump outlets; such placements are a source of errors in the RTD measurement via periodic microphonic effects depending on the flux in the pipeline imposed by the nearest pump [24].

All necessary detectors were fixed, collimated and cabled to a data acquisition system (DAS) and a laptop, while the background radiation level was measured prior to the radiotracer preparation and injection. Software CAESAR II v.12 (Hexagon PPM, Stockholm, Sweden) was used to treat the data obtained by the DAS.

2.3. Radioisotope

$^{99m}$Tc was selected as a radiotracer in this experiment since it remains stable at the prevailing process conditions, having a short half-life and low gamma energy. The half-life of $^{99m}$Tc is 6 h and gamma energy is 140 keV [25]. $^{99m}$Tc is commonly used as an aqueous phase tracer. About 62 mCi radioisotope $^{99m}$Tc was eluted in the form of sodium.
pertechnetate solution from $^{99m}$Tc generator (Nuclear Research Centre Rež, Czech Republic), and 0.1, 0.3, 0.5, 0.7 and 1.0 cm$^3$ of the solution having activities of 0.22, 0.68, 1.12, 1.58 and 2.25 mCi, respectively, were used in experiments under the same operating conditions.

2. Methodology and data treatment

Instantaneous impulse (approximated as Dirac) input of radiotracer into the system was effected for each of the measurements performed and can be seen as a black peak in Fig. 2. Since the peak is sharp and well defined, the former statement stands. The number of counts in 0.1 s intervals was measured at the outlet by the three collimated NaI(Tl) detectors positioned at different output positions. Detector 1 monitored the quality of the injection.

The RTD data were always corrected for the background. The RTD can be calculated from the measured concentration at the outlet as a function of time, $c(t)$. From the pulse experiment the RTD, $E(t)$, can be determined using Eq. (2). Using the first and second moments of the RTD curve, the mean residence time $\tau$ (Eq. 3) and its standard deviation $\sigma$ (Eq. 4), respectively, can be calculated.

$$E(t) = \frac{c(t)}{\int c(t)dt} \quad (2)$$

$$\tau = \mu_1 = \frac{\int t\cdot c(t)dt}{\int c(t)dt}$$

$$\sigma^2 = \mu_2 - \mu_1^2 = \int t^2\cdot c(t)dt - \frac{(\int t\cdot c(t)dt)^2}{\int c(t)dt} \quad (3)$$

By modifying Eqs (2–4) into sums throughout the whole concentration distribution time-interval, the mean residence time and its standard deviation were calculated directly from the raw dataset.

The flow rate was calculated as average value of $\tau$ from all 3 detectors. Hence $\Delta t$ (transient time) in the Eq. (1) represents the mean value of $\tau$ from all 3 detectors.

Another way to determine MRT was the RTD software developed at CEA Saclay, France. Within the software, plug flow with axial dispersion model was used. Its default concentration distribution curve is described by the equation:

$$c(t) = \frac{1}{2\sqrt{\pi t^3}} e^{-\frac{Pe(t-\tau)^2}{4\tau^2}} \quad (5)$$

where $Pe$ is the Péclet dimensionless number.

2. Safety features

Radiological safety has been monitored throughout the experiment. Personal protective equipment (PPE) and injection tools were collected into radiation waste plastic bags, and these sealed bags were transferred to the radiation bunker until the radioactivity does not exceed the permissible limit. After 15 half-life cycles, the waste can be disposed as normal waste. Dose received by the operator did not exceed the levels set by the The French Alternative Energies and Atomic Energy Commission (CEA).

3. RESULTS AND DISCUSSION

Figure 2 shows typical data set measured at all 4 detection points, where broadening of the responses could be noticed indicating slight axial dispersion.

First, cumulative growth fit (Boltzmann) was used to find the correct MRT (or $\tau$) value, as shown in Fig. 3. Deviation was spread throughout the whole calculation via the error propagation law. At the end, a combined uncertainty
containing both statistical and systematical errors was calculated along with relative bias of the flow rate from the set value of 10 dm$^3$ min$^{-1}$ (set on the flow meter). Results calculated using this method are summarized in Table 1.

Figure 4 shows, on the other hand, the RTD software solutions of the experimental data for axial dispersed plug flow for $^{99m}$Tc volume of 0.7 cm$^3$ having the activity of 1.58 mCi.

Results obtained by the RTD software applied to the data measured at the detection point D3 are presented in Table 2, which shows average flow rate, absolute bias from 10 dm$^3$ min$^{-1}$, standard deviation and relative deviation of the calculated flow rate.

![Figure 2. RTD experimental curves recorded by detectors placed at the radiotracer inlet and at the outlet at 50, 70 and 100 cm from the injection point for the $^{99m}$Tc volume of 0.7 cm$^3$ and activity of 1.58 mCi](image)

![Figure 3. MRT ($\tau$) dependency on time showing cumulative focusing of the final MRT value throughout the whole RTD-curve range. Parameter A2 of the Boltzmann fit shows the MRT value when time approaches infinity. The iteration step equals to the measurement window of 0.1s](image)
Table 1. Flow rates with combined and relative deviations for different ⁹⁹mTc injection volumes. Calculated via sum approximation of raw data measured at D3.

<table>
<thead>
<tr>
<th>V / µL</th>
<th>Q_{exp} / dm³ min⁻¹</th>
<th>σ Q_{exp} / dm³ min⁻¹</th>
<th>σ / %</th>
<th>Bias, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>11.29</td>
<td>0.27</td>
<td>2.39</td>
<td>12.89</td>
</tr>
<tr>
<td>300</td>
<td>10.96</td>
<td>0.11</td>
<td>0.99</td>
<td>9.57</td>
</tr>
<tr>
<td>500</td>
<td>10.87</td>
<td>0.16</td>
<td>1.48</td>
<td>8.68</td>
</tr>
<tr>
<td>700</td>
<td>10.89</td>
<td>0.11</td>
<td>0.98</td>
<td>8.90</td>
</tr>
<tr>
<td>1000</td>
<td>10.92</td>
<td>0.18</td>
<td>1.67</td>
<td>9.18</td>
</tr>
</tbody>
</table>

Figure 4. RTD software solutions to the experimental data measured at D3 for axial dispersed plug flow for ⁹⁹mTc volume of 0.7 cm³ having the activity of 1.58 mCi: E(t) denotes the signal at the inlet, S(t) represents the signal measured at the outlet, E(t)*H(t) represents calculated response of the model to a given signal E(t), H(t) is the impulse response of the model and * denotes the convolution operation. Pe = 9.77

Table 2. Flow rates with standard and relative deviations for different ⁹⁹mTc injection volumes. Calculated via the RTD software from data measured at D3

<table>
<thead>
<tr>
<th>V / µL</th>
<th>Q_{calc} / dm³ min⁻¹</th>
<th>σ Q_{calc} / dm³ min⁻¹</th>
<th>σ / %</th>
<th>Bias, %</th>
</tr>
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<tbody>
<tr>
<td>100</td>
<td>11.09</td>
<td>0.13</td>
<td>1.19</td>
<td>10.88</td>
</tr>
<tr>
<td>300</td>
<td>11.13</td>
<td>0.10</td>
<td>0.94</td>
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<tr>
<td>500</td>
<td>11.04</td>
<td>0.12</td>
<td>1.05</td>
<td>10.38</td>
</tr>
<tr>
<td>700</td>
<td>11.07</td>
<td>0.14</td>
<td>1.23</td>
<td>10.66</td>
</tr>
<tr>
<td>1000</td>
<td>11.11</td>
<td>0.14</td>
<td>1.28</td>
<td>11.06</td>
</tr>
</tbody>
</table>

By comparing results shown in Tables 1 and 2, it can be seen that bias is in general lower when using the summation method, which may point to better validity of the results (excluding the 100 µL injection) as the flow meter was set to 10 dm³ min⁻¹. The relative standard deviation, on the other hand, is generally lower for the RTD software processed data giving a hint of higher precision for this method. It is also evident that RTD software results are more consistent. Nevertheless, summation approximation using RTD curve moments can give similar data without the need of using a special software.

However, Tables 1 and 2 also show that both methods provide the best results in the range of 300–700 µL injection volume. Such an outcome might point to a slight output dependency on the injection volume. Injection volume, i.e. the input activity, being too low may lead to higher relative standard deviation, especially when using manual summation calculation. Such phenomenon can be simply understood based on the statistical character of radioactive decay, where lower activity leads to a higher relative error of measured number of counts (N), which is defined by the Gaussian character of N as sqrt(N)/N. Hence, lower activity leads to higher statistical errors of a quantity calculated by the measured count rate. Of course, this error further propagates to any quantity derived from N (such as MRT).
In agreement with the basic statistical law, the relative standard deviation of the calculated flow rate decreases with increasing the injection volume (activity). However, at 1000 µL injection volume, i.e. the maximum volume used in this experiment, the relative standard deviation increases again, which was not expected. This effect might be explained by detected irregularities caused by dead-time triggering around the top of the RTD peak area, since there is no deviation from the pulse input, and all of the input signals can be approximated as Dirac function. The determination of two borderline values of the calculated quantity may suggest the existence of an optimum injection volume range.

One of the most important findings of this study is that after a detailed literature lookup, the authors did not find another study that investigates the change of standard deviation of the calculated flow rate with the injection volume and activity of used radiotracer. Hence, this study was conducted for the first time.

4. CONCLUSIONS

Experiments on a pilot-scale flow system have been performed using \(^{99m}\text{Tc}\) as a radiotracer in the form of pertechnetate ion (\(^{99m}\text{TcO}_4^-\)) to measure flow rate for flow meter calibration. The data were evaluated by using a plug flow with axial dispersion model applied by a RTD software as well as by the summation approximation (Eqs 2–4). The results of the study showed that there might be an optimum injection volume range of the tracer for each specific application. The optimum range in this study was determined as 300 to 700 µL. In this range, the relative standard deviation was around 1% for both calculation methods, which is lower than that obtained with other tested injection volumes. Bias of the determined flow rate was lower for the summation approximation, resulting in ca 9% in the area of interest. In the case of RTD software, bias values were between 10 and 11%. The results of the study may help in injection volume optimization and provides a guide on data processing without the RTD software, as the results obtained by both methods were in great agreement. This investigation of the change in standard deviation of the calculated flow rate as a function of the injection volume and activity of the radiotracer used, is the first according to the literature.

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SAŽETAK

Optimizacija zapremine radioaktivnog obeleživača pri merenju zapremskog protoka u zatvorenim cevovodima

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(Naučni rad)

U hemijskim procesima od suštinskog značaja je tačno definisan i precisan protok. Merenja brzine fluida su važna za karakterizaciju kvaliteta protoka tečnosti u sistemima. Ova studija se fokusira na određivanje zapremskog protoka i standardnog (relativnog) odstupanja izmerenih vrednosti u cilju kalibracije konvencionalnih merača protoka korišćenjem metode radioaktivnog obeleživača. Merenja potrebna za kalibraciju merača protoka izvedena su na pilot postrojenju korišćenjem Tehnecijuma-99m (99mTc) kao radiotrejera u obliku pertechnetatnog jona (99mTcO4-). Izmereni podaci su analizirani, a preciznost eksperimentalne postavke je ispitivana pomoću dva različita pristupa: upotrebljavanje softvera za određivanje raspodele vremena zadržavanja (eng. residence time distribution, RTD) koga je razvila Međunarodna agencija za atomsku energiju i zbirne aproksimacije neobrađenih podataka. Po prvi put je utvrđena varijacija standardne devijacije izračunatog protoka u odnosu na zapreminu i u odnosu na aktivnost ubrizganog radioaktivnog obeleživača. Model protoka sa aksijalnom disperzijom je korišćen za simulaciju izmerenih RTD krivih i istraživanje dinamike protoka vode. Rezultati studije su pokazali mogućnost in situ kalibracije merača protoka sa relativnom greškom manjom od 1%. Takođe su pokazali malu zavisnost preciznosti izlaznih rezultata od količine ubrizganog obeleživača, kao i slične rezultate za manuelnu obradu i specijalizovanu obradu podataka upotrebom RTD softvera.

Ključne reči: radioaktivni obeleživač; RTD; tehnicijum-99m; merač protoka; kalibracija; hemijska industrija.