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Validation of transient time method to calibrate oil flow meters in closed conduits using ^{123}I as the radiotracer

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Abstract. To assure the credibility of commodity transfer operations in the oil and gas industry, calibrated flow meters are used to quantify the movement of fluids in the pipelines. The purpose of this paper is to validate the transient time method (TTM) to calibrate oil flow meters installed in restricted areas, using the ^{123}I-labelled oil as a radiotracer. Traditionally, as proposed by the standard ISO 2975-7:1977 [1] for experiments in an aqueous medium, the TTM is employed by positioning two detectors at separate locations. However, in industrial plants, it is not always possible to install detectors at the distances recommended by the ISO 2975-7. The method proposed in this paper uses four scintillator detectors separated one from each other by 0.30 m and three injections containing 5.0 ml of ^{123}I-labelled oil. The experiments were carried out in an oil flow rig with a turbulent flow profile. The results have reached an uncertainty which is lower than 1.0%.

Keywords: Calibration flow meters • Oil flow rates • Iodine-123 • Transient time method

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Introduction

The oil industry production chain is extensive and complex, and in most of the stages, the transportation of oil and oil products is carried out using pipelines. The royalties and taxes are calculated by measuring the oil volumes using certified flow meters, and these devices must be periodically calibrated. The result of these measurements must comply with the specific quality standards [2–4] and should be according to the guidelines specified by the official governance [5–7]. In Brazil, it is necessary to comply with regulations [8–10] that have been established by the National Petroleum Agency (ANP), which imposes uncertainty levels lower than 1.0% in the measurements.

For aqueous fluid flow in pipelines, the flow measurement is regulated by the ISO 2975-7:1977 [1] standard, which recommends the transient time method (TTM) [11–15] to calibrate the flow meters installed in pipelines, because this method is accurate and less invasive. The objective of this work was to validate the TTM to calibrate oil flow meters installed in closed conduits using ^{123}I-labelled oil as the radiotracer. The TTM is designed to be applied in locations with restricted space, as in the off-shore oil platforms.

Traditionally, the use of radiotracers is based on three fundamental aspects [16]: simplicity, low

cost, and accuracy [17], and according to Clayton [18], the right radiotracer should have the following characteristics:

i. Solubility and hydrodynamic characteristics compatible with those of the primary fluid;
ii. Short half-life coherent with time to be measured; and
iii. Since the volume of radiotracer injected is small, the gamma energy must be high enough to be detected outside the duct.

The experiments were carried out in an oil flow rig built with PVC ducts (3/4 in internal diameter). It consists of two sections: the first one contains the oil reservoir, the pumping system, a calibrated flow meter, the injection system, and the second part is the measure section used to install the detectors. It is 16.0 m long. The fluid used in the study was the Lubrax Essential 20W50 API SJ/SJ oil and an electronic system regulates and keeps the oil flow constant during the experiments.

Oil labelling with iodine-I23

The labelling process begins with an aqueous solution of sodium iodide (2.0 ml, 74 MBq) labelled with ^{123}I (E = 159 keV and $T_{1/2}$ = 13.2 h). It uses iodine monochloride (ICl) as the radioiodine carrier [19, 20]. ICl is a polarized molecule which is used in electrophilic substitution reactions to attach the radioactive iodine to the double bonds present in the oil molecule. ICl is more effective in those substitution reactions than other iodine compounds because it is easily ionizable and highly reactive.

A sample (100.0 μL) of the initial Na^{123}I was collected (labelling stage LS-0) to monitor the labelling process. Its activity was measured and used as a reference value. In each labelling stage, samples were collected (100.0 μl), and the gamma activity was measured using a dosage calibrator.

The labelling process starts with the production of ICl with the reduction of potassium iodate (KIO_3) in an acid medium. In a separation funnel, 5.0 ml of KI (0.1 N) was mixed with water (5.0 ml) and 10.0 ml of concentrated HCl acid (4 N). After this, the Na^{123}I radioactive solution (LS-0 labelling step) is added to the system, and KIO_3 (0.1 N) is gradually added. In the concentrated HCl medium, the iodate is reduced to iodine monochloride, as in the equation:

$$(1)\qquad KIO_3 + HCl + 2\,KI + Na^{123}I \rightarrow 3\,^{123}ICl + 3\,KCl + 3\,H_2O$$

The balance of the Eq. (1) is controlled using 1.0 ml of carbon tetrachloride (CCl_4) as the colour indicator. There are two phases in the separation funnel: an aqueous phase, which is slightly yellow, and a purple CCl_4 phase. This CCl_4 colour indicates that there is still iodine in the solution, and the stoichiometric balance was not achieved.

When the stoichiometric balance of the Eq. (1) is reached, the ICl is extracted from by adding 2.0 ml of ethyl ether and vigorously shaking the system for 5 min. After that, maintain the system in rest until the separation of the two phases occurs. This procedure is repeated twice (LS-1A and LS-1B labelling stage) to ensure that the radioiodine is extracted.

The organic phase is transferred to a new container with 5.0 ml of regular oil and shaken for 2 min. After that, the oil is heated at 40°C for 30 min to remove the ethyl ether (LS-2 labelling stage).

To ensure that radioactive iodine is bonded to the oil molecules and not free in the medium, the labelled oil is washed with water: 5.0 ml of labelled oil is introduced by slowly shaking in 50.0 ml of water for 5.0 min, and the two-phase mixture was separated by centrifugation. This operation was repeated twice, and the gamma activity was measured (LS-3A and LS-3B labelling step).

The gamma activity was measured using a dosage calibrator (Capintec CRC-15W), and the results for the different label steps are in Table 1. The aqueous samples are identified by W and the oil samples by O.

The residual activity at WLS-1B shows that 95.0% of the initial activity was transferred to the organic phase because, after two extraction steps, only 5.0% of radioiodine was measured in the aqueous phase. The activity measured at stages OLS-3A and OLS-3B reinforce the inference that the radioiodine was bonded to the oil molecule, because if there were any free radioiodine in the oil, it would be transferred to the aqueous phase.

The oil labelling process is better assessed by measuring the iodine number [21, 22] that determines the number of existing double bonds in the oil molecules. After the labelling process, the number of double bonds between carbon atoms in the oil molecules is reduced, and this value is certified by the change in the iodine number [18, 19].

Before the labelling process, the number of iodine measured was equal 66.37 ± 0.43; after the labelling step 2, the number of iodine of OLS-2 was measured again, and the result was 6.75 ± 0.62. This reduction proves that the iodine is bonded to the oil.

Due to the high yield of the labelling, 85%, the method can be used not only in lab tests, but also in providing services in industrial plants. To apply it in experiments that require high activity, it is necessary only to vary the activity of the initial NaI sample without changing the concentrations of the reagents used in every labelling stage. Another favourable characteristic is that the radiotracer used is produced especially for medical purposes, and this condition allows many groups of radiotracers in countries that import this radiopharmaceutical to use this method to label oil products to use them for industrial applications.

Table 1. The yield of ^{123}I oil labelling process

Sample	WLS-1A	WLS-1B	OLS-2	WLS-3A	WLS-3B	OLS-3A	OLS-3B
Yield (%)	7.81	5.12	88.74	0.44	0.38	85.46	84.92

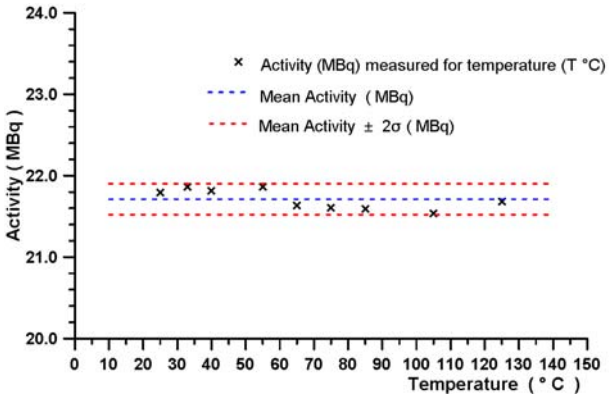

Fig. 1. Influence of temperature on the labelled oil stability.

One important aspect is the stability of the radiotracer in different conditions. An experiment was done to evaluate the temperature effect on the labelled oil: an Erlenmeyer with 50.0 ml of labelled oil was gradually heated, and dry air was pumped to remove any free volatilized iodine. During the test, the temperature varied from 20°C to 130°C, and for each temperature value, 100.0 µl of labelled oil was removed, and the activity was measured. Figure 1 shows the results, and it is clear that the labelled oil is not affected by the temperature increase since the activity remains stable.

Further, to accurately measure the flow rate, it is essential to prove that labelled oil reacts neither with the walls of the pipeline nor with the decants inside the pipeline. A device was built to confirm that the labelled oil maintains the same characteristic as regular oil. This decice is a cylinder with 0.80 m of the same duct used in the pipeline, which is closed at the base and covered with a removable plug in the top used to fill the unit with oil. Figure 2 shows the device mounted with three collimated NaI scintillating detectors (1 inch × 1 inch) to register the radiotracer signals at the upper, middle, and base of the device.

Fig. 2. The apparatus used to study the radiotracer stability.

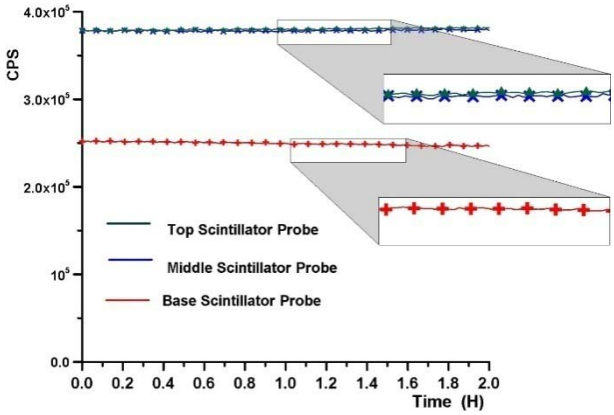

Fig. 3. Three scintillator probes registering the gamma count at the base, middle, and top sector of the test device with 74 MBq of labelled oil.

In a 500 ml glass Becker, 235.0 ml of regular oil and 5.0 ml of labelled oil were slowly shaken for 10 min to avoid air bubbles, and then transferred to the measuring device; thereafter, the cap is closed. The three detectors registered the number of gamma photons for 2 h. If any precipitation occurred, the base counts would increase, or if the labelled oil moved toward the surface, the top counts would increase. However, if the counts remain stable during the test, it is guaranteed that the labelled oil maintains the same properties as regular oil. Figure 3 shows the results measuring the gamma activity for 2 h.

In Fig. 3, the top signal is the same as the middle one, and the base signal is smaller than the others because the solid angle was different in that position. The result confirms that labelled oil presents the same characteristics as the fluid used in this flow measurement study.

Flow meter calibration in oil tubes using the transient time method

According to ISO 2975-7:1977, the recommended condition to apply TTM to measure water flow in closed conduits is that the flow should be uniform and in a turbulent regime, with Reynolds number higher than 5000 [1], the radiotracer injected as a fast pulse, and the two detectors which are used separated by a considerable distance.

However, in space-restricted areas, the amount of space which is available to install detectors is limited. The developed method proposes to install four detectors in a straight portion of the pipeline; each one is separated from the other by 0.30 m, and three radiotracer injections are used for a single flow rate measurement (fast pulse). For each set of two scintillator detectors, the oil flow Q is calculated by:

$$Q = A \cdot \frac{L}{\mathrm{TAU}} = A \cdot v = \frac{\pi D^2}{4} \cdot v \tag{2}$$

where: A – area of pipeline cross section, L – the distance between two scintillator detectors, TAU – transient time, v – mean flow velocity, D – internal diameter of the pipe.

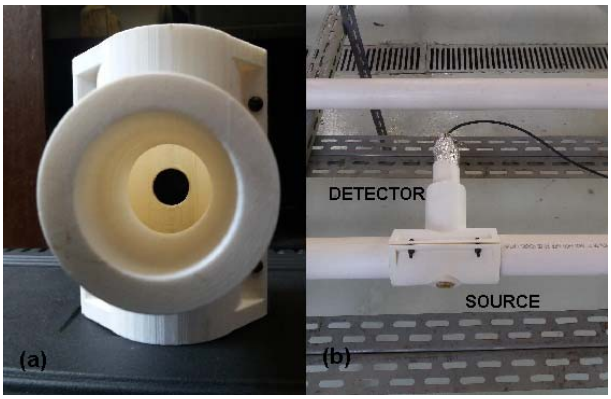

Fig. 4. A top view of the scattering device in (a) and (b) is used to analyse the internal pipe wall.

TTM requires that the pipeline must be full. Thus, it can only be applied in a measure section where there are no dense material deposits or air bubbles inside.

A scattering device was built to scan the measure section. It is a scintillator detector, and an ^{241}Am source is mounted each one in one side of the duct and used to measure the gamma transmitted radiation in two directions: vertically and horizontally, perpendicular to the flow direction. Figure 4a shows the device, and Fig. 4b shows the scattering device installed on the experimental oil flow rig.

Table 2 shows the counts for scanned measure positions, and the results have shown that there is no evidence of dense material or air bubbles. There is a 2.0% difference between the transmitted radiation registered in vertical and horizontal directions; this difference is due to the back-scattering contribution because, in this position, the pipeline was mounted near a solid metal structure.

Using TTM to measure a flow rate is relatively simple: it is done by installing the detectors, injecting an ideal radiotracer, and calculating the TAU between the two measuring points. The difficulty increases when it is necessary to measure the flow rate according to the official limits [1], especially in industrial plants, where restrictions to install the detectors exist.

Since the radiotracer movement inside the duct depends on the flow velocity profile, the correct positions to install the detectors give the best result. These positions are selected based on the injection point, the fluid velocity, tracer dispersion, and the pipeline layout.

The method uses four detectors and measures the TAU in each position. This configuration results in six independent values for the flow velocity. Thus, after repeating the procedure for the three injections, we will have eighteen values that must be combined using a proper statistical method to calculate the flow rate.

In all injections, the radiotracer solution (5.0 ml, $1.5{\cdot}10^8$ Bq) was injected as fast pulse using a bypass system at 5.0 m upstream of the measurement section to minimize any tail contribution.

In all measurements, a lead collimator with a rectangular slit (1.0 cm) was used to block the frontal face of each scintillator detector, and the lateral face was shielded using lead bricks (5.0 cm). The data acquisition system was programed to register the radiotracer signal with a sample rate equal to 20 Hz.

Results

For each injection, after calculating the six mean flow velocities, the Dixon test (95%) [23] was applied to verify if there is an outlier and to remove any significant different value from the data set. When the data set is homogeneous, the mean V_M and the associated uncertainty $u(V_M)$ are calculated.

The same procedure is repeated for the other injections, and the F-test for multiple comparison procedures was applied to determine whether there exists a significant difference among the three injections. If there is no difference among the treatments, the flow rate Q, and the uncertainty $u(Q)$ are calculated considering the three injections [24]. The results for the reference values, RV, equal to 800 LPH, 950 LPH, and 1000 LPH are shown, respectively, in Tables 3–5.

Table 6 shows for each reference value (RV), the measured flow rate (Q), the relative uncertainty (rv%), and the error calculated comparing the measured flow rate with the reference value measured using the calibrated flow meter installed in the oil flow rig.

Table 2. Pipeline scan in the measurement positions using a ^{241}Am gamma source

	Total gamma-ray counts			
	Position D1	Position D2	Position D3	Position D4
Vertical	8188 ± 90	8194 ± 90	8209 ± 91	8175 ± 90
Horizontal	8002 ± 89	8000 ± 89	8001 ± 90	7990 ± 89

Table 3. Mean flow velocity (in cm/s) for injections T1, T2, and T3, and RV = 800 L/h (Reynolds number = 8085)

Injection	D1D2	D1D3	D1D4	D2D3	D2D4	D3D4	V_M
T1	76.95	78.36	77.53	77.59	76.87	77.87	77.53
T2	76.89	77.99	78.09	77.87	78.22	77.96	77.84
T3	76.83	78.39	77.60	77.99	78.14	78.19	77.86
			Mean velocity			77.74 ± 0.27 cm/s	
			Oil flow rate			797.72 ± 6.57 L/h	

V_M – mean value.

Table 4. Mean flow velocity (in cm/s) for injections T1, T2, and T3, and RV = 950 L/h (Reynolds number = 9600)

Injection	D1D2	D1D3	D1D4	D2D3	D2D4	D3D4	V_M
T1	93.16	90.07	92.24	91.85	93.87	91.12	92.04
T2	93.11	91.02	91.02	91.12	92.23	92.85	92.01
T3	93.07	90.47	91.92	93.09	93.89	90.86	92.50
			Mean velocity			92.18 ± 0.26 cm/s	
			Oil flow rate			944.84 ± 7.56 L/h	

V_M – mean value.

Table 5. Mean flow velocity (in cm/s) for injections T1, T2, and T3, and RV = 1000 L/h (Reynolds number = 10 107)

Injection	D1D2	D1D3	D1D4	D2D3	D2D4	D3D4	V_M
T1	97.26	96.95	98.89	97.14	97.78	97.08	97.52
T2	96.86	98.22	97.13	96.99	98.08	97.68	97.49
T3	98.46	97.21	98.09	97.39	97.01	97.95	97.69
			Mean velocity			97.57 ± 0.31 cm/s	
			Oil flow rate			1001.14 ± 8.16 L/h	

V_M – mean value.

Table 6. Relationship between flow measurement data

RV ± 0.25% (L/h)	*Q* (L/h)	rv%	Percent error
800	797.72 ± 6.57	0.824	0.28
950	944.84 ± 7.56	0.800	0.54
1000	1001.14 ± 8.16	0.815	0.11

Conclusions

By using the iodine-labelling method, it was possible to produce the radiotracer labelled with ^{123}I with high yield (85%) without changing the labelled oil characteristics. The tests show that the radiotracer produced remains stable for oil temperatures between 20°C and 120°C.

For the three experiments, the flow rate results correspond to a relative standard uncertainty which equals 0.8%. According to Metrological Technical Regulation INMETRO 64/2003 [10], the method corresponds to Class 1 and can be applied for calibrating the flow meters in purchase/sale transactions.

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