The influence of optical density change on measurement error of refractive index in flowing liquid

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Abstract. The article discusses the physical processes in a flowing fluid that affect the change in its optical density. Changes in the refractive index and its errors in single-component and multicomponent media for different fluid flow regimes are investigated. A new model has been developed for calculating error of refractive index, considering the non-uniformity of optical density in a stream liquid. A technique is proposed for selecting a refractometer model for reliable monitoring the state of liquid environment.

1. Introduction

The deterioration of the ecological state of water bodies, the introduction of automatic processes in the production of different products and other factors require a person to constantly improve means of monitoring the state of condensed matter [1–8]. The greatest difficulties arise in monitoring the state of liquids flowing in pipelines of various diameters. Especially if the measurements to determine the liquid environment state should not make irreversible changes in its physical structure and chemical composition [8-16]. Therefore, the greatest preference when conducting such measurements is given to contactless methods [1-6, 9-12, 14-21]. Among them, the greatest advantages compared with others, has a method based on the use of the phenomenon of refraction [22-31]. The control of the medium state in the methods using the refraction phenomenon is carried out by the measured value of the refractive index n or the position of the light-shadow border [22, 26-28]. Usually for scientific research or to control the state of the technological process, for example, in the pharmaceutical industry, the values of n with an error of no higher than $10^{-4}$ should be measured [1, 17, 18, 23, 24].

There is factor that has been little studied, this factor has a significant effect on the measurement error n. This factor is the change in the optical density of the liquid. Especially, this factor is pronounced in turbulent flows. This greatly limits the use of refractometers for process control. Research is needed to develop a way to compensate for the effect of a change in optical density on the measurement error n of a flowing fluid. The research results and the method of calculating the change in the optical density of the flowing fluid to the value of n will increase the knowledge in the description of refraction.

2. The effect of optical density on error measurement of the refractive index

To measure the refractive index of a flowing fluid, various types of refractometers are used [17, 20, 23–31]. Generally, preference is given to differential type refractometers, in which compensation schemes are used to reduce the effect of various negative phenomena on the measurement error n [1, 23, 29-32]. The use of compensation schemes in the case of laminar flow of a liquid makes it possible to obtain measurement errors of n not worse than $10^{-4}$. When measuring n in a turbulent fluid flow, the
measurement error n increases. Therefore, the reasons that cause this process should be considered in more detail.

In the study of the liquid flow, the use of compensation devices in a refractometer is possible only if the device is built according to a differential scheme [29-32]. The developed designs of differential type automatic flow refractometers for measuring n use the Anderson difference prism method with a differential cell. The basic design of the differential (difference) cell consists of two tanks [29-31]. According to one container (flow cell) the investigated liquid moves. Its refractive index n_1 must be controlled. In the second tank (closed cell) is a liquid medium with a known refractive index n_c.

In fig. 1 shows a block diagram of a differential cuvette (technical name - cuvette transducer (CT)) and the course of laser radiation in it.

![Figure 1. Propagation of laser beams in a differential cuvette.](image)

The flowing fluid enters the flow cell 2. With the fluid flowing rapidly in the cell, the fluid layers are mixed. To make this process less intense, the diameter d_T is made much smaller than L_1. In addition, the entrance to the flow cell and the liquid outlet from it are located opposite each other. Our studies have shown that this leads to the formation of a stagnant zone (in Fig. 1 this is part of the flow cell next to the designation of the angle α). With long-term measurements, especially medical suspensions, this can lead to the formation of foreign structures hazardous to human health (for example, when controlling the process in pharmaceutical companies).

Therefore, we proposed a different principle for locating the position of pipelines, which is shown in Fig. 1. In this case, the mixing of the liquid layers is more significant than in the previously used CT schemes, but the probability of the formation of a stagnant zone in the CT is much less.

Consider the mechanism of the occurrence of the error, which is caused by fluctuations in the optical density of the measured liquid. According to the Lambert-Beer law, the light value at the output of a CT, if it is filled only with a comparative fluid, is determined by the following formula:

\[ E_0 = \frac{\Phi_0}{2ab} 10^{-D_0} = \frac{\Phi_0}{2ab} 10^{-K_0L_0}, \quad (1) \]

where \( \Phi_0 \) – input light output CT; \( K_0 \) - specific optical density ratio of the reference fluid; \( L_0 \) – optical base CT; \( D_0 \) - optical density of a comparative fluid.

Consider the central beam B. For a simpler consideration, we choose the ideal case when the beam is almost not refracted on the partition between the flow cell and the closed cell (angle α is small). Then the central beam (B) passes the path l_1, in a cuvette filled with a comparative fluid, and the path l_2 in the flowing fluid. The specific coefficient of optical density, which can vary in the flowing fluid, is denoted as \( K_x \). Extreme rays A and C pass through various paths in the measuring and comparative fluids and
are attenuated differently, which causes an imbalance of the measuring circuit (MC) not caused by the displacement Δx and the occurrence of an error. The distances \( l_1 \) and \( l_2 \) traveled by an arbitrary beam lagging behind the middle beam B by the value of \( y (-a \leq y \leq a) \), in comparative and controlled fluids, are determined by the expressions:

\[
\begin{align*}
  l_{1y} &= l_1 - y \cdot \tan \alpha \\
  l_{2y} &= l_2 + y \cdot \tan \alpha
\end{align*}
\]

(2)

where \( l_1 + l_2 = L_0 \).

Different distances traveled by the rays in liquids with different specific coefficients of optical density, lead to ambiguous absorption of the light beam in the CT along its width 2a.

Consequently, the illuminance \( E \) depends on the y coordinate (the laser beam propagates along the y axis), taking into account the variations in optical density \( \Delta K = K_2 - K_0 \):

\[
E(y) = \frac{\Phi_0}{2a} \cdot 10^{-((K_0 l_1 + K_2 l_2) - \Delta K \tan \alpha)}
\]

(3)

In accordance with the previously discussed theories to determine the error of the signal at the output of the MC expression (3) can be represented as follows:

\[
(\Delta z_1)_0 = L b \left( \int_{-a}^{0} E(y) \, dy - \int_{a}^{0} E(y) \, dy \right) = L \frac{\Phi_0}{2a} \cdot 10^{-((K_0 l_1 + K_2 l_2) - \Delta K \tan \alpha)} \times \left( \int_{-a}^{0} 10^{-\Delta K \tan \alpha} \, dy - \int_{a}^{0} 10^{-\Delta K \tan \alpha} \, dy \right)
\]

(4)

Substituting relation (3) into (4) and taking into account (1) and (2), we can obtain the following expression for the error signal provided that the refractive index of the liquid does not change:

\[
(\Delta z_1)_0 = L \frac{\Phi_0}{2a} \cdot 10^{-((K_0 l_1 + K_2 l_2) - \Delta K \tan \alpha)} - \frac{\chi[2.3025 \Delta K \tan \alpha] - 1}{2.3025 \Delta K \tan \alpha}.
\]

(5)

Relation (5) determines the error at the output of the MC of a differential automatic refractometer with a differential measurement circuit at zero. It should be noted that when \( \Delta K \to 0 \) in the formula (5) uncertainty arises, when revealed by the rule of L'Hôpital we get \( (\Delta z_1)_0 = 0 \), what corresponds \( E(y) \to E_0 \). This is confirmed by experiment..

The complex expression (5) for absolute error makes it difficult to use for practical purposes. The transition to relative values allows us to simplify the error formula, which can be represented by the following expression:

\[
\delta(\Delta n) = \delta_1(0) + \delta_2(\Delta n),
\]

(6)

where \( \delta_1(0) \) – characterizes the offset "zero" when the deviation of optical density from the normalized value; \( \delta_2(\Delta n) \) – functional term associated with the measured error value \( \Delta n \).

For addend \( \delta_1(0) \) in (6), which characterizes the offset, the following formula was obtained:

\[
\delta_1(0) = 0.0993 \frac{\Delta L^2 \Delta d}{l \Delta n_{\text{max}}},
\]

(7)

where \( \Delta L \) – half of width light beam; \( l \) – distance from the differential cell to the photodetector; \( \Delta d = d_0 - d_x \) – change in the optical density of the measured liquid \( d_x \) relative to the initial (unreinforced) value \( d_0 \).
For another term in (6) the following formula is obtained:

$$\delta_2(\Delta n) = -\frac{1.15\tan^2\alpha(\Delta n)^2}{\Delta n_{\text{max}}} \Delta d,$$

where $\alpha$ - refracting angle of the cuvette; $\Delta n = n_f - n_c$ – the difference in refractive index measured ($n_f$) and comparative ($n_c$) liquid flows.

Our studies have shown that for refractometers with a compensation measurement scheme, formula (6) takes the following form

$$\delta(\Delta n) = \delta_1(0),$$

In this case, the measurement error $n$ in these refractometers is determined only by the offset of the “zero” caused by the change in optical density ($\Delta d \neq 0$).

3. The results of calculations of the effect of changes in optical density on the error in measuring the refractive index

For different values of the difference between the refractive indices $\Delta n$, changes in optical density, width of the light flux and other typical parameters of refractometers, the values of the components of the absolute error $\delta(\Delta n)$ were calculated. The results of the calculations are presented in tables 1 and 2.

### Table 1.

| $\Delta n_{\text{max}}$ | $\Delta d$, см | $\Delta L$, см | $l=30$ см | $l=40$ см | $l=50$ см |
|-------------------------|----------------|--------------|-----------|-----------|-----------|
| 0,001                   | 0,001          | 0,1          | 0,031     | 0,023     | 0,019     |
|                         | 0,01           | 0,1          | 0,124     | 0,092     | 0,076     |
|                         | 0,1            | 0,1          | 0,311     | 0,233     | 0,187     |
|                         | 0,2            | 0,2          | 1,244     | 0,932     | 0,748     |
|                         | 0,1            | 0,2          | 3,110     | 2,333     | 1,866     |
|                         | 0,2            | 0,1          | 12,440    | 9,992     | 7,464     |
| 0,01                    | 0,001          | 0,1          | 0,003     | 0,002     | 0,002     |
|                         | 0,01           | 0,1          | 0,012     | 0,004     | 0,004     |
|                         | 0,1            | 0,1          | 0,031     | 0,23      | 0,19      |
|                         | 0,2            | 0,2          | 0,124     | 0,920     | 0,076     |
|                         | 0,1            | 0,2          | 0,311     | 0,233     | 0,187     |
|                         | 0,2            | 0,1          | 1,244     | 0,932     | 0,748     |
| 0,1                     | 0,001          | 0,1          | 0,003     | 0,003     | 0,002     |
|                         | 0,01           | 0,2          | 0,012     | 0,008     | 0,008     |
|                         | 0,1            | 0,1          | 0,031     | 0,023     | 0,019     |
|                         | 0,2            | 0,2          | 0,124     | 0,092     | 0,076     |

### Table 2.

| $\Delta n_{\text{max}}$ | $\Delta d$, см | $\alpha$, ° | $l=30$ см | $l=40$ см | $l=50$ см |
|-------------------------|----------------|-------------|-----------|-----------|-----------|
| 0,001                   | 0,001          | 45          | 0,003     | 0,004     | 0,006     |
|                         | 0,01           | 60          | 0,009     | 0,012     | 0,018     |
|                         | 0,1            | 70          | 0,023     | 0,030     | 0,045     |
For series-connected links of the differential refractometer, the values of \( \sum(\Delta n) \) were calculated. For the calculations used the following formula:

\[
\sum(\Delta n) = \frac{\delta_2(\Delta n)}{\delta_1(0)} = 1.2 \left( \frac{\tan \Delta n_{\text{max}}}{\alpha} \right) k^2,
\]

where \( k = \frac{\Delta n}{\Delta n_{\text{max}}} \leq 1 \). \( \Delta n_{\text{max}} \) – the maximum possible difference between the refractive indices of the liquid and the reference fluid.

The results of calculations \( \sum(\Delta n) \) are presented in table 3.

**Table 3.**

| \( \Delta n_{\text{max}} \) | \( k \) | \( \alpha \), ° | \( \sum(\Delta n) \), % | \( l=30 \text{ см} \) | \( l=40 \text{ см} \) | \( l=50 \text{ см} \) |
|-----------------------------|-------|----------------|-----------------|----------------|----------------|----------------|
| \( 0,001 \)                 | 0,1   | 45             | 0,001           | 0,002           | 0,003           |
| \( 0,01 \)                  | 0,01  | 45             | 0,003           | 0,006           | 0,009           |
| \( 0,1 \)                   | 0,1   | 45             | 0,008           | 0,015           | 0,023           |
| \( 0,001 \)                 | 0,5   | 45             | 0,027           | 0,048           | 0,075           |
| \( 0,01 \)                  | 0,01  | 45             | 0,081           | 0,144           | 0,225           |
| \( 0,1 \)                   | 0,1   | 45             | 0,203           | 0,360           | 0,563           |
| \( 0,001 \)                 | 0,5   | 45             | 0,108           | 0,192           | 0,300           |
| \( 0,01 \)                  | 0,01  | 45             | 0,324           | 0,576           | 0,900           |
| \( 0,1 \)                   | 0,1   | 45             | 0,810           | 1,440           | 2,250           |
Analysis of the calculation results (Tables 1-3) allows us to draw the following conclusion. With increasing $\Delta n$, the sensitivity of the measurement error $n_f$ to a change in optical density decreases. This fact is confirmed by the experiment.

4. Conclusion

The experimental results obtained by us confirmed the model proposed by us for calculating the error in measuring the value of $n$, which is related to the nonuniformity of optical density in a flowing fluid, and showed its universality. The model can be used for both stationary and flowing fluid.

Using this model allows us to estimate the magnitude of the measurement errors $n$ for various liquids and determine the feasibility of using a differential type refractometer to monitor their status. Or use other types of measuring devices to monitor the state of the liquid environment, since the measurement error $n$ does not satisfy, for example, the requirements of the experiment.

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