The universal optical method for condition control of flowing medium

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Abstract. In the article peculiarities of determination of dissolved substances concentration change in flowing liquid medium by measuring its refractive index with refractometer are considered. The measuring results of different media refractive indices are presented. The investigations of their refractive index dependencies on different factors (temperature, concentration, etc.) are carried out. The capabilities of refractometer applications for flowing medium state control at light-shadow borders displacement from refracted laser radiation are showed.

1. Introduction

Experimental and theoretical studies of liquid medium streams are one of the typical problems of physics [1-3]. The research results are important for improving the design of various meters of physical quantities of the liquid medium flow, developing new methods for measuring them and monitoring the state of the flowing medium [3-7].

The most difficult is to study the flow of aggressive and dangerous media (for example, benzene, heptane, concentrated sulfuric acid, etc.), as well as in cases when sterility is required (for example, biological solutions, medical suspensions, part of food products, etc.) [1, 3, 6]. Measurements in most of the considered flows of liquid media must be carried out by methods in which there is no direct contact between the measuring elements of the instrument and the medium under study. In some cases, small contact of the measuring elements with the flow of the medium is possible (for example, optical materials, etc.) [7-9]. But the following condition must be satisfied. With long contact of such elements with the medium under study, they should not alter its structure and chemical composition (sterility is ensured by installing such instruments on the pipeline for measurement).

Today there is a constant increase in competition in the food industry, which requires the manufacturer to produce high quality products. On the other hand, it is necessary to optimize the costs of its production. Reducing the cost of manufacturing products in large food production industries is provided mainly through the automation of the technological process, ensuring its continuity and control at all stages of production. Therefore, in contrast to other cases, additional requirements are imposed on instruments for monitoring the state of the flowing medium (production products, for example, juice, oil, wine, etc.) that are a part of the overall system for controlling the technological process of production.
In addition to reliable operation for a long period of time, real-time monitoring of the environment, simplicity of construction and relatively low cost (they are quite numerous - more than 10, the control of the medium should not change its taste qualities, etc. The latter is especially important, as experience shows in the production of liquid food products, control of its quality must be carried out at various stages of production for the rapid elimination of failures and violations of technology.

Most of the measuring instruments successfully used to solve problems of contactless monitoring of the flowing medium in various areas of industrial production and services could not satisfy the additional requirements. The exception was only nuclear magnetic spectrometers, but they are expensive, even with a wholesale purchase (more than 10 devices) [3, 10, 11]. Therefore, the search for solutions to this problem is quite urgent and in demand.

One of the possible solution is to use a refractometer. Using signals based on measuring the refractive index $n_m$ of the flowing medium (for example, juice, etc.), a refractometer mounted directly on the pipeline can monitor and control the production process.

### 2. Refractometer for controlling liquid stream condition

The method of refractometry makes it possible to study stationary and flowing stream of liquid media by measuring their refractive indices $n_m$. Using refraction ratios, according to the results of measurements $n_m$, chemical compounds can be qualified, physicochemical parameters of various media can be determined, and structural and quantitative analysis can be carried out [8, 12, 13].

Today a large number of refractometers of various designs have been developed for measuring $n_m$ in steady-state and in the flowing state of liquid media with high accuracy (the measurement error in most cases does not exceed 0.3%). But they are designed, especially flowing, to solve local problems. For example, one of the best is the CM-800 alpha-SW refractometer - salt meter designed to measure the NaCl salt concentration in water with an error of 0.1% in continuous mode. Refractometer models that allow measuring $n_m$ in the range from 1.33000 to 1.55700 with an error of 0.05% (for example, PRM-100 alpha (ATAGO, Japan)) are quite expensive.

There are various chemical compounds (acids, alcohols, etc.), in which the values of $n_m$ are less than 1.33 (at $T = 293 \, K$). For example, hydrochloric acid HCl has the values of $n_m = 1.2540$, methyl alcohol CH3OH — 1.3296, etc. With increasing T, the refractive index of the medium $n_m$ decreases insignificantly (in general, less than 10% with increasing T up to 373 K). These media are used in technological processes in various industries (food and chemical, etc). Their condition must be controlled by contactless methods because, for example, HCl is an aggressive medium.

Using the described above refractometers does not allow solving this problem. These devices are mainly designed to control the production of various fertilizers (for example, aqueous solutions of urea, etc.), technical liquids (for example, aqueous solutions of ethylene glycol and propylene glycol that are used at airports to remove icing from the hull of the aircraft, etc.) etc.

Our research has shown that the expansion of the measurement range of $n_m$ by a refractometer is primarily related to the parameters of its optical part. Therefore, we made an optical system based on the newly developed sapphire prism design. Its structural scheme is shown in figure 1.

![Figure 1. Structural diagram of the optical part of the refractometer](image-url)
In the developed design of the optical part of the refractometer, the source of laser radiation is set up in such a way that its rays after passing through the optical system reach the boundary of the prism, which contacts the flowing medium at different angles. Part of the rays with angle of incidence greater than the critical angle $\alpha_c$ (figure 1) completely reflects from the inner surface of the prism and, emerging from it, forms the light part of the image on the photodiode ruler. The critical angle $\alpha_c$ is determined by the following relation:

$$\alpha_c = \arcsin \left( \frac{n_m}{n_p} \right)$$

where $n_p$ is the refractive index of the material from which the prism is made; $n_m$ is the refractive index of the flowing medium.

The remaining rays, the angle of incidence of which is less than the critical $\alpha_c$, are partially refracted and pass into the solution, partially reflected and form the dark part of the image on the photodiode ruler (figure 1). In a small range of angles from $\alpha_c$, an interface between light and shadow is formed. The contrast of this interface between light and shadow depends on the relation between the refractive indices of the material (1) from which the prism and the investigated medium are made, as well as the parameters of the laser radiation (wavelength $\lambda$, divergence angle $\theta$ and spatial coherence length $L_k$).

On the basis of our studies, the following has been established. The coefficient of reflection of laser radiation from the prism-medium boundary to the value of the critical angle changes monotonically (the Fresnel formula). In a small range of incidence angles, before reaching the value of $\alpha_c$, the reflection coefficient increases sharply (with an increase of one order of magnitude higher than before). For laser radiation rays incident on the prism-medium interface at angles $\alpha \geq \alpha_c$, the reflection coefficient has a value of 1. If the center of the beam pattern of a laser falls on the interface between two media at an angle $\alpha_c$, then the light-shadow boundary contrast ratio is maximal. The presence of a peak in the intensity of the detected laser radiation along the length of the photodiode line (photodetector) is due to two factors. The distribution of intensity in the rays of laser radiation forms a cone (with a maximum in the center). For a coefficient of 1, this distribution does not change. A sharp change in the reflection coefficient before reaching the critical angle of incidence appears. This change makes the intensity maximum distinct.

In case of a drop in the center of the radiation pattern at a different angle, the light-shadow boundary widens (the contrast decreases). This can possibly lead to an error in measurements. That is why the refractometer needs a preliminary calibration before measurements.

When the composition of the medium changes, its refractive index $n_m$ changes and the position of the light-shadow boundary recorded by the photodiode ruler is shifted. We propose to monitor the state of the flowing liquid using this method, in contrast to previously used in refractometers based on the measured value of the refractive index $n_m$.

Another distinguishing feature of the developed refractometer from other models is the use of a new design of a prism made of leucosapphire in its optical system which geometric configuration and dimensions are shown in figure 2.

**Figure 2.** Geometric shape and dimensions of the prism
The research showed that the calculated and implemented geometry of the prism provides the possibility of the refractometer to monitor the state of the flowing medium in the range of $n_m$ from 1.2246 to 1.6120. This result was achieved, because this design of the prism allowed us to use laser radiation with a plane angle of the radiation pattern of about 22.6° (after the optical system) and to move the position of the semiconductor laser along the base of the prism within 8 mm. To cover this working range, we used a photodiode line TSL1406R with a photo-sensitive layer length of 38.77 mm. This ruler is placed at a small distance from the base of the prism (4-5 mm) and can move freely along it. In the design of the model TSL1406R, a plate with an enlightenment is placed in front of the photosensitive layer. On the inner side of this plate a special layer is applied, which makes unimportant the influence of radiation reflection effects inside the photodiode ruler on the measurement result. Between the prism and the photodiode ruler there are no focusing optical elements. Parasitic reflections can occur only between the outer edges of the protective plastic of the photodiode ruler and the prism.

Let $I_0$ be the maximum intensity of the radiation incident on the plate. The reflection coefficient of the plate is less than 3%. The reflection coefficient of sapphire for laser radiation incident from air is not more than 7%. The value of the parasitic intensity received secondarily on the plate is less than 0.0021·$I_0$. The experiments have shown that with a sharp change in the reflection coefficient at the prism-current interface, the phenomenon considered does not significantly affect the contrast value of the light-shadow boundary.

Besides, the expansion of the $n_m$ measurement range was ensured by using a cone shaped gasket in the measuring probe of the refractometer with the prism (figure 2). This gasket makes the influence of the vignetting effect of the laser beam on the faces not significant as compared to the previously used in the refractometer prism of trapezoidal geometry with an annular gasket. The conical gasket also provided greater sealing reliability, which is very important in a pipeline with fast fluid flow of high pressure. To process information from the photodiode line, we have developed a program that allows us to determine the refractive index values $n_m$, the relative concentration $N_m$, as well as the temperature coefficient $dn_m/dT$ after the preliminary calibration of the instrument.

The calibration of the refractive index scale $n_m$ was carried out using the normalized output signal $A$ from the photodiode array:

$$A = \frac{\sum_{i=1}^{256} A_i}{A_{max}}$$

(2)

where $A_{max}$ is the maximum value of the illumination signal from the photodiode ruler, $A_i$ is the amplitude of the signal from the photodiode cell element, and $i$ is the element number.

To test the performance of the refractometer we used reference aqueous solutions of ethylene glycol and 1,2 propylene glycol with different relative concentrations of $N$ from 0 to 100%.

3. The results of experimental studies of the flowing liquids

Figure 3 shows the results of measuring different refractive indices $n_m(T)$ for liquid aqueous solutions of ethylene glycol and 1,2 propylene glycol with different relative (mass) concentration $N$. The solid line in figure 3 shows a linear interpolation of these dependencies.
Figure 3 (a,b). Temperature dependences of the refractive index $n_m$ of aqueous solutions of ethylene glycol (a) and 1,2 propylene glycol (b). For graphics 1, 2, 3, 4 and 5, the following concentration of N in % corresponds to (a): 0.00; 5.22; 14.85; 31.40; 100.00, for (b): 0.00; 4.59; 13.41; 27.59; 100.00.

Analysis of the figure 3 dependencies shows that, at $T$ of about 333 K, the dependency $n_m(T)$ is linear and coincides with the linear interpolation, which was calculated for each graph. Also, for each graph (figure 3), we calculated the mean square convergence $\Delta n_m$ of interpolation dependence calculations and experiments. Table 1 presents the results of our calculations.

Table 1. Linear interpolation formulas for the variation of $n_m$ from the temperature $T$ for different concentrations of N aqueous solutions of ethylene glycol and 1.2 propylene glycol and the values of $\Delta n_m$.

| Aqueous solution of ethylene glycol | An aqueous solution of 1.2% propylene glycol |
|------------------------------------|---------------------------------------------|
| N, %                              | $n_m$, rel.un. | $\Delta n_m$, rel.un. | N, %                              | $n_m$, rel.un. | $\Delta n_m$, rel.un. |
| 0.00                              | 1.3357 – 0.00010339·T | 0.00155 | 0.00 | 1.3357 – 0.00010339·T | 0.00155 |
| 5.22                              | 1.3407 – 0.00013007·T | 0.00087 | 4.59 | 1.3415 – 0.00012569·T | 0.00088 |
| 14.85                             | 1.3506 – 0.00015162·T | 0.00085 | 13.40 | 1.3509 – 0.00014340·T | 0.00083 |
| 31.40                             | 1.3683 – 0.00019586·T | 0.00081 | 27.59 | 1.3682 – 0.00018155·T | 0.00078 |
| 100.00                            | 1.4370 – 0.00027520·T | 0.00061 | 100.00 | 1.4385 – 0.00030048·T | 0.00065 |

We have established that the error of linear interpolation for the concentration $N \leq 5\%$ is about 0.00155. For values $N > 5\%$, the values of $\Delta n_m$ do not exceed 0.00088. The obtained results coincided within the measurement error of previously obtained results in [14-16] using the “Expert pro” refractometer (the medium under study is in a stationary state).

Experimental studies have shown that in the developed refractometer with the use of the new prism design, an exceptional feature that is inherent in some models of optical measuring instruments has been implemented. It is connected with the fact that the optical scheme of the refractometer is based on the use of reflection and light transmission only within the working prism. Therefore, factors such as the transparency of the flowing medium, the presence of light scattering insoluble elements in it (for example, pulp in juice, etc.), gas bubbles, as well as the flow rate do not affect the measurement results, unlike other methods (except NMR), used to monitor the stream of liquid medium. The light-shadow boundary shifts only when $n_m$ changes. This allows us to accurately determine the presence of changes in the composition of the flowing medium, for example, on the concentration of N or T. Figure 4, as an example of the refractometer operation, studies of flowing aqueous solutions of various media at a temperature $T = 293.1\, K$. 
Figure 4. Dependence of the refractive index of aqueous solutions of some substances on concentration: 1 - gelatin, 2 - sodium glutamate, 3 - sucrose, 4 - ethyl alcohol

An analysis of the obtained experimental results showed that the variation of $n_m$ from $N$ for most media is nonlinear, in contrast to the dependence of $n_m(T)$. At low concentrations of $N$ (not more than 10%) and $T$ no more than 293 K for some media, the device allows measuring small changes in $n_m$ on $N$. This allows using it to monitor the state of a large number of liquid media during their flow in the pipeline.

Figure 5 shows, for example, the experimental dependencies of the change in the relative density of the aqueous solution in Brix units (Brix is the most common calibration scale for refractometers, expresses the concentration of a solution of chemically pure sucrose in distilled water in mass percent) with the addition of sugar in various concentrations from temperature $T$. As the reference point for the change in the relative concentration, the value of $\Delta N = 0$ at $T = 293$.

Figure 5. Temperature corrections for the concentration of aqueous solutions of sucrose: 1 - 10%, 2 - 30%, 3 - 60%

Experimental studies have established that the most important factor that can make errors in determining the state of the medium is $T$. To account the effect of change in $T$ of the investigated flowing medium on the results of monitoring its state in the flowing liquid, a temperature sensor is installed in the pipeline in front of the prism. Based on the readings of this sensor, a correction is taken into account.
in the displacement of the light-shadow boundary by means of the gradient tables. The discrepancy between the amount of boundary displacement and the data of the table means that there are additional changes in the state of the medium that are not related to changes in the temperature T. Similarly, the temperature factor must be taken into account when controlling the state of the medium in terms of nm [8, 14-16].

4. Conclusion
The research showed that the use of the optical system developed by us with a new design of a sapphire prism makes it possible to monitor the state of the flowing liquid medium with \( n_m \) from 1.2246 to 1.6120 with an error of 0.1%. We have established that in order to improve the accuracy of monitoring the state of the flowing medium, it is necessary for each variety to select the appropriate wavelength and laser radiation power at which the light-shadow border contrast will be maximum. The obtained result allows using the developed refractometer to control the state of the flowing medium of bromobenzene \( n_m = 1.5601 \), which is used to prepare reagents for various chemical reactions. Also this refractometer can be used in the medical industry, e.g. to control the state of the phenylhydrazine \( n_m = 1.6105 \) in the manufacture of medicines (e.g. antipyrine, amidopyrine). Today other types of instruments are used for their control.

Moreover, it has been discovered that to ensure a stable operation of the refractometer to monitor the state of the flowing liquid medium, regardless of its flow rate, it must be installed on the vertical section of the main line. Sometimes the pipeline is filled with an incompletely flowing liquid in the technological process of production, especially food liquid products. (In the technological process of production, especially food liquid products, there are quite often situations where the pipeline is filled with an incompletely flowing liquid.) The contact of the upper face of the prism with the measured medium will be absent. This circumstance will not allow the measurement of various parameters of the flowing medium and the control of its state with the required accuracy.

The restriction in the application of the refractometer is also the diameter of the pipeline through which the liquid medium flows. Because the flow at the borders of the pipeline is much less than in the center, and if a refractometer is placed on the vertical section of the pipeline, the prism should be the upper edge partially flooded into the steam of liquid medium. The smaller the diameter of the pipeline, the smaller the geometric dimensions of the prism, the smaller the detectable dynamic range of the light-shadow boundary and the measuring range \( n_m \).

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