



EMERGING TRENDS IN OPTICAL SENSOR APPLICATIONS FOR AQUATIC ENVIRONMENT

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Abstract

Multi-channel polarization optical technology is increasingly used for prompt monitoring of water systems. Optical devices during the assessment of water quality determine the intensity of light through the studied aquatic environment. Spectrophotometric devices measure the spectrum of weakening of light through the aquatic environment. Spectroellipsometric devices receive spectra in vertical and horizontal polarizations. The presented article develops an adaptive optical hardware and image system for monitoring water bodies. The system is combined. It consists of 2 parts: 1) automated spectrophotometer-refractometer, and 2) adaptive spectroellipsometer. The system is equipped with a corresponding algorithmic and software, including algorithms for identifying spectral curves, databases and knowledge of spectral curves algorithms for solving reverse problems. The presented system is original since it differs from modern foreign systems by a new method of spectrophotometric and spectroellipsometric measurements, an original elemental base of polarization optics and a comprehensive mathematical approach to assessing the quality of a water body. There are no rotating polarization elements in the system. Therefore, this makes it possible to increase the signal-to-noise ratio and, as a result, improve measurement stability and simplify multichannel spectrophotometers and spectroellipsometers. The proposed system can be used in various water systems where it is necessary to assess water quality or identify the presence of a certain set of chemical elements.

Keywords: Monitoring; Aquatic environment; Polarization optics; Water object; Pollutants; Spectral images; Classification; Identification

1. Introduction

The assessment of physicochemical parameters (PCP) of aquatic systems is the subject of many studies aimed at creating technical and algorithmic means for measuring and processing data on the state of the aquatic environment. Remote technologies have acquired particular relevance in recent years, allowing us to obtain operational information about water bodies and characterized by high productivity. The greatest effect is achieved

when using multi-channel remote sensing systems, when, through the use of the necessary set of algorithmic tools, it is possible to significantly increase the reliability of the interpretation of observational data and successfully solve assessment problems of water quality. Work in this direction is being carried out at the Institute of Radio Engineering and Electronics named after V.A. Kotelnikov RAS. The works formulate the basic principles of the integrated use of mathematical and technical means for solving problems of hydrophysical monitoring ^[1]. According to these principles, the procedure for synthesizing an automated data processing system for multichannel water quality measurements includes the creation of a set of hardware, algorithmic, model and software tools for collecting and analyzing information, taking into account the levels of its reliability and completeness ^[2,3].

The aquatic environment, as studies by many authors have shown, requires the collection of a large amount of data and, as a rule, laboratory analyzes for its assessment. The latter circumstance sharply worsens the functional characteristics of the monitoring system. The conducted studies excluded the stage of laboratory analysis from the procedure for assessing the quality of the aquatic environment, moving it to the beginning of the procedure for adopting the identification algorithm. This feature of the developed methodology is a fundamental difference between the proposed approach to assessing the physicochemical characteristics of water bodies from the developed approaches by other authors. In this case, the advantages of multichannel measurements in the optical and microwave regions of the spectrum are used, which, as shown by experimental measurements and theoretical models, creates the possibility of using highly efficient information technologies to solve problems of classification and identification of water bodies. The implementation of the methodology makes it possible to reduce the volume of observations and thereby increase the efficiency of the monitoring system.

The spectral composition of the radiation emerging from water carries information about the absorbing and scattering substances contained in the water. This is the physical basis of remote methods for analyzing the composition of natural waters. Knowledge of the spectral distribution of the brightness coefficient of waters with a known chemical composition makes it possible to determine some of the ingredients of the unknown composition of waters in the presence of characteristic features of the brightness of the water body under study. Recently, spectral-polarization-optical methods and corresponding equipment for real-time research have been intensively developed: multichannel polarization spectrophotometers; spectropolarimeters; spectral ellipsometers and dichrometers; and refractometers. The use of effective polarization state modulators and multichannel analyzers in modern polarization-optical devices, as well as the development of programming methods, determine their high technical characteristics.

The joint use of technical and software tools for operational monitoring of the aquatic environment is poorly developed due to the complexity of synthesizing a comprehensive monitoring system. Particularly difficult are the tasks of combining algorithmic support with the level of information support of the monitoring system. The urgent task of environmental monitoring requires the development of compact precision polarization optical instruments for express analysis of liquid media. At the same time, the effectiveness of solving multiparameter problems is largely determined by the sensitivity and accuracy of devices, their versatility, and the ability to use a wide spectral range. Spectral measurements in the aquatic environment create an informative basis for the use of modern methods and algorithms for recognizing and identifying pollutants in this environment.

2. Materials and methods

2.1 Spectrophotometry and spectroellipsometry methods

Spectrophotometry and spectroellipsometry are optical techniques that use changes in the polarization state of light through reflection and refraction to characterize surfaces, interfaces, thin films in physics, and liquid

solutions in hydro-chemical studies in situ and in real time. Spectrophotometry and spectroellipsometry methods are used for non-destructive testing of the chemical and physical characteristics of solid materials and liquid solutions. These methods are based on recording the effects of optical polarization that occur when a light wave is reflected or deformed as a result of its interaction with the object under study. Diagnostics of liquids allows you to assess the concentration of chemical substances dissolved and suspended in the liquid, as well as identify stains of contaminants on the surface of the water.

Spectrophotometry is a method of research and analysis of substances based on measuring absorption spectra in the optical electromagnetic region. radiation. The basis of spectrophotometry is a physicochemical method for studying solutions and solids, based on the study of absorption spectra in the ultraviolet (200–400 nm), visible (400–760 nm) and infrared (> 760 nm) regions of the spectrum. The main dependence studied in spectrophotometry is the dependence of the absorption intensity of incident light on the wavelength. The result is a discrete spectrum, which, using traditional techniques, is converted into a continuous spectrum, which is visualized and analyzed. Optical spectral analysis is characterized by the relative ease of implementation, the absence of complex sample preparation for analysis, and the small amount of substance (10–30 mg) required for the analysis of a large number of elements.

The traditional approach to analyzing spectral images of solutions is based on cluster analysis. First, pattern recognition is trained by forming a cluster space. Then, based on the fact that the spectral image enters a given cluster, a decision is made on the composition of this solution. This fairly simple method works well for one-component solutions. In more complex cases, interpolation methods and more complex procedures are used. One of these procedures will consist of the following.

To solve the problem of recognizing the structure and content of chemical elements in an aqueous solution using a spectrophotometer, at the initial stage a database of standard samples is created. To create it, measurements were taken of samples of one-component and two-component solutions of various chemical elements.

Forming a database of standards for samples of aqueous solutions requires the creation of a formalized indicator for each sample. For this purpose, the Department of Computer Science of the Institute of Radio Engineering and Electronics of the Russian Academy of Sciences carried out measurements of one-component and two-component solutions with a resolution of 5% of the maximum possible concentration of a chemical element. This concentration was determined at the expert level. To develop an informative indicator of each spectrum, the encoding method adopted for signal recognition in radio engineering was used. For each spectrum, a vector indicator-image was created and clusters were formed.

Multichannel spectroellipsometry methods are among the most informative and sensitive methods for studying solid, liquid and gaseous objects. For example, the thickness and optical constants of monolayer coatings on the surface of liquids or the presence and structure of molecules of optically active substances in solutions are reliably measured.

Spectroellipsometers are designed for real-time measurements of the spectra of ellipsometric parameters Ψ and Δ ($\tan\Psi$, $\cos\Delta$) with subsequent transition, within the framework of a specific physical model of the structure under study, to the spectra of optical constants and geometric parameters (thicknesses of films and transition layers, degree of surface roughness, etc.).

The ability to accurately and quickly determine the spectra of optical constants of various materials and film thicknesses in the range from 1 nm to several microns has determined the widespread use of spectral ellipsometry in electronics, chemistry and electrochemistry, physics, biology, and medicine. Recently, spectroellipsometers have found interesting applications in the non-destructive determination of the critical dimensions of nano-

electronic structures. Laser ellipsometers with lateral resolution better than $0.1\ \mu\text{m}$ are appearing. The use of spectral and laser ellipsometers as sensitive sensors is also of interest. The spectroellipsometer described below does not contain moving polarization elements, unlike widely used multichannel spectroellipsometers with rotating polarizers, analyzers and compensators, which allows for increasing the sensitivity of the spectroellipsometer in the absence of vibration noise.

The inverted geometry of the spectroellipsometer makes it possible to reduce the influence of background illumination during measurements with weak signals.

It should be noted that increasing the sensitivity and long-term stability of polarization-optical devices is achieved by using various modulators of the polarization state (photoelastic, Faraday, acoustooptical, etc.). In multichannel spectroellipsometers with rulers and photodetector arrays, the optimal use of rotating polarization elements (polarizer, compensator or analyzer). At the Institute of Radio Engineering and Electronics named after V.A. Kotelnikov RAS is developing a new approach to spectroellipsometry—spectroellipsometry with binary modulation of the polarization state, using an original binary polarization modulator, effectively replacing known expensive polarization elements^[3–8].

In the spectroellipsometer with binary modulation (SEBM), radiation with two orthogonal polarization states is applied sequentially in time to the sample under study. The radiation beam reflected from the sample is either divided by a Wollaston^[3–5] prism into two orthogonally polarized beams with azimuths A and $A + 90^\circ$, which are simultaneously directed to two photodetectors, or passes through a polarization device that selects polarization sequentially in time with azimuths A and $A + 90^\circ$, and arrives to a photo detector (linear or matrix of photodetectors).

The absence of moving polarization elements and the compactness of a SEBM of the polarization state provide a high signal-to-noise value and allow it to be used for measurements in field conditions. Currently, SEBM has a wide spectral range, high sensitivity and operational reliability. The use of these spectroellipsometers for diagnosing various water bodies has shown high efficiency.

2.2 Spectrophotometric and spectroellipsometric tools

Figure 1 shows the one developed at the Kotelnikov IRE RAS combined spectrophotometer and spectroellipsometer. The LED spectrophotometer (LSP) and LED spectroellipsometer (LSE) consist of two parts. The top part contains a spectrophotometer, and the lower part is a spectroellipsometer.

LSP is designed for training, which is a procedure for measuring the spectral characteristics and simultaneous independent measurement of the content of chemical elements in the aquatic environment. As a result, a database of standards is formed in the knowledge base, a comparison which provides a solution to the problem of identification. In particular, such a comparison can be realized within the framework of the calculation of the average square deviation of the measured spectral image of an object from the standards held in computer memory. The LSP software provides various algorithms for solving this problem, among which there is discriminant and cluster analysis.



Figure 1. A combined Led spectrophotometer and spectroellipsometer.

LED Spectroellipsometer includes:

- a polarizer that converts a linearly polarized light flux into elliptical polarization;
- an analyzer that evaluates the parameters of the ellipse;
- power supply that supplies voltage according to the selected operating mode of the spectroellipsometer;
- light source with known spectral characteristics;
- fiberglass cable;
- broadband filters.

LES measures in real time the spectra of two ellipsometric angles Ψ and Δ ($0 \leq \Psi \leq 90^\circ$, $0 \leq \Delta < 360^\circ$; $\tan \Psi$, $\cos \Delta$) [3–8]. Based on these parameters, by solving the inverse problem of spectroellipsometry within the framework of a specific physical and mathematical model of the object (structure) under study, a transition to geophysical and geochemical parameters is realized (content of a chemical element, types of spots on the water surface, temperature, salinity, transparency, etc.).

Three measurement modes are possible:

- 1) In the first case, the measurement procedure is limited to taking a sample of the solution and placing it in a special cuvette with known optical characteristics. This limitation is leveled by the fact that here at the output we obtain two spectra reflecting the ellipsometric angles that determine the ratio of the complex amplitude refractive indices of light for two polarizations. Possible distortions of the spectra may occur in the case of uncontrolled changes in the characteristics of the cuvette.
- 2) Direct measurements of the integral flux of scattered and refracted light by lowering the light guide and receiver into a liquid medium. In this case, external interference is excluded.
- 3) Use of sunlight reflected from the water surface. In this case, the level of illumination of the surrounding space is simultaneously recorded to calibrate the obtained spectra. The level of instability of the recorded spectra is determined by the instability of the illumination of the surrounding space during the signal registration time (fractions of a second).

In any mode of measuring the optical characteristics of the test object η at the output of the spectro ellipsometric system according to the basic equation $\rho = r_p/r_s = \tan \Psi \exp(i\Delta)$, where Ψ and Δ are the ellipsometric angles that determine the ratio of the complex amplitude reflection coefficients r_p and r_s for p and s polarizations, two spectra are obtained [3–6]:

- 1) $S_\Psi(\lambda, \eta)$ —spectral distribution of the tangent of the spectroellipsometric angle Ψ .
- 2) $S_\Delta(\lambda, \eta)$ —spectral distribution of the cosine of the spectroellipsometric angle Δ .

Spectral curves $S_{\Psi}(\lambda, \eta)$ and $S_{\Delta}(\lambda, \eta)$ are functions of the optical characteristics of the diagnosed object η . Based on these spectra, one can estimate the physical or chemical parameters of an object by solving the inverse problem of spectroellipsometry.

The algorithmic and functional capabilities of LES make it possible to implement a training procedure for spectral pattern recognition. In this case, a database of spectral standards is formed in LES, and the inverse problem of spectroellipsometry is solved by searching this database for spectra close to the new spectrum obtained. The accuracy of solving the problem depends on the method of assessing this proximity.

2.3 One method for solving the inverse problem

For the experiment, 6 solutions of copper sulfate (CuSO_4) of various concentrations were prepared, namely 5%, 10%, 15%, 20%, 25%, 30%. By mixing two solutions in certain proportions, you can get a third solution of intermediate concentration from the list above. To obtain the proportions of two solutions, it is necessary to solve the inverse problem by comparing the readings of the spectrophotometer of the linear combination of two solutions with the readings of the instrument of the third solution. The problem is described by a system of linear equations:

$$Ax = B$$

(1)

where the vector corresponds to the spectrum of the third solution. We will solve (1) by the Cramer method. Consider a combination of 3 solutions: 5-20-10, i.e. we need to mix two solutions with a concentration of 5% and 20% in order to obtain a

concentration of 10%. We will choose two reference channels: $\lambda_1 = 380 \text{ nm}$ and $\lambda_2 = 525 \text{ nm}$.

Channel selection can be optimized according to various criteria. The simplest is the choice of the maximum channel separation. But since the channels $\lambda = 660 \text{ nm}$ and $\lambda = 780 \text{ nm}$ with are extremely noisy, we will exclude them from consideration and stop at the $\lambda = 525 \text{ nm}$ channel. The 365 nm channel, being the leftmost channel, was omitted to align the pair of channels to the center of the frequency range.

$$A = \begin{vmatrix} 0.76683 & 0.21598 \\ 0.77462 & 0.39918 \end{vmatrix} \quad b = \begin{vmatrix} 0.59794 \\ 0.75117 \end{vmatrix}$$

$$\det A = 0.13881, \det A_x = 0.076448, \text{ then } x = \det A_x / \det A = 0.55077$$

$$\det A_y = 0.11284 \quad y = \det A_y / \det A = 0.81299$$

Here the designation is accepted: $x_1 = x$, $x_2 = y$. How to evaluate the quality of a popular result? Here, too, different optimality criteria are possible, for example, the normalization error. In our case, it is 0.36376. The second criterion is to minimize the deviation of the obtained proportion of the first solution with a concentration of 5% from the theoretical estimate. In our case, this deviation is 0.11589. In this method, we did not take into account the normalization condition.

Let us now consider how the normalization condition affects the quality of the solution. Under these conditions, solving the second equation for, corresponding to the channel $x = 525 \text{ nm}$, we obtain $x = 0.93753$, which, according to the second criterion, gives an error equal to 0.27087, which is more than 3 times greater than the corresponding error. But by sorting through all the channels, we can easily find the optimal one, in this case it is a channel with a wavelength $\lambda = 380 \text{ nm}$, and the corresponding error is 0.02674, which is an order of magnitude less than the error obtained with the first method according to the second criterion. Even the worst 470 nm channel has an error comparable to the error of the first method, namely 0.38460. Solving the problem for the 380 nm and 365 nm channels using the 1st method, we get an error for both criteria even greater than for the previous pair of channels. Thus, it is obvious that the optimization of the decision procedure should still be carried out taking into account the normalization condition, especially since the enumeration of options in this case is much smaller. Another thing is when the answer to the problem is unknown, i.e. the third solution has an unknown intermediate concentration. In this case, it is necessary to sort through all possible pairs of spectral channels and perform optimization according to the first criterion.

We have studied all 20 combinations of three solutions and obtained optimal error estimates for the second criterion. The worst channels and their corresponding error estimates were also found.

We will distinguish between combinations of two types: general (**Table 2**) and symmetrical (**Table 3**) combination. We call a combination of 3 solutions symmetrical if the proportions of two of them in the third solution are equal.

These combinations in our experiment are:

1. 5-15-10; 2. 5-25-15; 3. 10-20-15; 4. 10-30-20;
5. 20-30-25; 6. 15-25-20

The remaining combinations of solutions will be called combinations of the general. Let us now present the final optimization results with the indication of the minimum and maximum errors and the corresponding channel.

Table 2. General combinations.

View	Channel	Minimum	Channel	Maximum
5-25-10	365	0.01204	470	0.29281
5-20-10	380	0.02674	470	0.38460

5-20-15	380	0.01976	470	0.45743
5-25-20	525	0.07403	400	0.17518
5-30-10	365	0.00899	470	0.23067
5-30-15	400	0.01896	470	0.27361
5-30-20	470	0.00402	365	0.19830
5-30-25	525	0.00824	470	0.08216
10-25-15	525	0.00869	400	0.20545
10-25-20	525	0.15824	400	0.23776
10-30-15	415	0.00831	525	0.11976
10-30-25	470	0.02498	365	0.09608
5-30-20	525	0.19170	365	0.28458
15-30-25	525	0.04214	365	0.10876

Table 3. Symmetrical combinations.

View	Channel	Minimum	Channel	Maximum
5-15-10	470	0.00000	365	0.16476
5-25-15	365, 380, 470	0.00000	415, 525	0.00001
10-20-15	380	0.00000	365	0.00043
10-30-20	365	0.00000	400	0.00020
20-30-25	365	0.00000	380	0.00004
15-25-20	470	0.11649	400	0.34918

It can be seen from the tables that symmetrical combinations of solutions are the most stable when solving inverse problems. This is apparently due to the fact that the errors of symmetrical solutions have different signs and the same weights, which leads to their mutual cancellation when summed up to the 5th decimal place.

As for the accuracy of optical instruments, it is determined directly by the accuracy of the spectral values, as well as the reliability of the final results of diagnostics of aquatic environments. The spectrophotometer measures the spectral curve $S(\lambda)$, and the spectroellipsometer measures the spectral curves $S_{\Psi}(\lambda)$ and $S_{\Delta}(\lambda)$, respectively. Optical diagnostics of water systems is carried out directly by analyzing spectral curves using specific algorithms and models [8,9,11,14–17].

3. Results and discussion

3.1 Application of LSP

LSP was used to determine the hardness of water.

Water hardness is a combination of chemical and physical properties of water associated with the content of dissolved salts of alkaline earth metals, mainly calcium and magnesium. Natural waters contain calcium and magnesium sulphates and bicarbonates,

i.e. Ca^{2+} and Mg^{2+} cations, SO_4^{2-} and HCO_3^- anions.

LSP was tested when determining the concentration of different solutions of CaCl_2 and MgSO_4 for different concentrations. The results are shown in **Table 4**.

For LSP, a Pascal program was developed that implements solution identification algorithms based on the standard deviation and discriminant analysis procedures.

LSP was also used to determine the content of

Ca⁺⁺ and Mg⁺⁺ ions in drinking water. When comparing the results of analyses of drinking water with SES data, the discrepancy was 2–3%.

Table 4. Concentrations of different solutions of CaCl₂ and MgSO₄.

Solution CaCl ₂			Solution MgSO ₄
Conc. %	λ	Indication. LSP	
0.25	365	–0.000013833	0.000013665
	380	–0.000016537	0.000000078589
	400	–0.000015107	–0.0000088009
	415	–0.000033185	–0.000017852
	470	–0.000015903	0.00000034272
	525	–0.000041114	–0.000019528
	660	–0.000044677	–0.000022124
	780	–0.000062126	–0.000038044
0.5	365	0.000054099	0.00011439
	380	0.000040307	0.00010519
	400	0.000058027	0.00013196
	415	0.000057663	0.00016455
	470	0.000063689	0.00014422
	525	0.000069659	0.00019617
	660	0.000059451	0.00017979
	780	0.000048948	0.00019018
1	365	0.00016419	0.00015503
	380	0.00017495	0.00024768
	400	0.00022308	0.00025534
	415	0.00025486	0.00031383
	470	0.00023397	0.0002794
	525	0.0003413	0.00043667
	660	0.0003493	0.00043667
	780	0.00037174	0.00047805

3.2 Application of LSE

LSE was used to assess water quality in Lake Baikal and the Siberian rivers Angara and Yenisei. Heavy metals and oil spills were discovered both in Lake Baikal and in the Angara and Yenisei rivers along the course.

In many countries, there is an increase in the concentration of heavy metals and petroleum products along the Arctic coast ^[13,14]. The main reason for the increase in this pollution is mainly the Siberian rivers, including the Angara and Yenisei. These rivers contain large industrial cities such as Angarsk, Irkutsk, and Krasnoyarsk. LSEs

have been used to estimate concentrations of heavy metals and oil spills in waters above and below these cities. The results of these measurements are shown in **Table 5**.

Table 5. Heavy metal concentration (ppb) in the river water samples was assessed using the LSE.

Measurement location	As	Cd	Cr	Cu	Ni	Pb	Zn
Lake Baikal	4.18	0.33	6.7	13.6	14.8	1.36	24.3
r. Angara to the dam	6.59	0.47	8.7	15.3	16.5	2.13	21.8
r. Angara after the dam	12.8	1.15	9.8	16.1	20.2	4.07	59.3
r. Yenisei to the confluence of the r. Angara	11.4	0.87	10.4	19.4	21.4	5.72	41.9
r. Yenisei after the confluence of the r. Angara	9.23	0.68	13.8	15.6	19.8	6.87	18.6

Based on the measurement results, the following conclusions can be drawn. In the Angara River and in the Yenisei River, all heavy metals are unevenly distributed. This is the result of a fast current and the turbulence of these rivers, as well as due to the rocky bottom of these rivers. It can be noted, in particular, that at the bottom of the Angara there are low areas where heavy metals accumulate and are periodically washed out. To obtain a real picture of the water quality of the Angara and Yenisei, it is necessary to develop a regional model of the water balance of the system of these rivers. This model should take into account the sources of pollution entering this system with inflows and wastewater ^[10–14].

4. Conclusions

The technology discussed in this paper for the combined use of spectrophotometry, spectroellipsometry and detection and classification algorithms can be used in determining the concentration of aqueous solutions (including medical solutions), as well as diagnosing the quality of wastewater from industrial enterprises in real time.

Also, LSP and LSE can be made in the form of a portable device, with the help of which the operator can monitor the quality of water resources in real time without taking samples and conducting chemical analyses in the laboratory. In a stationary version, they allow monitoring of the dynamics of water quality in a stream, and when placed on a ship—measure water parameters throughout the entire route. The functions of LSP and LSE can be achieved by increasing the volume of standards in the knowledge base.

Author Contributions

Ferdinant Mkrtchyan conceived the study design, developed the models, and drafted the manuscript; Vladimir Soldatov and Maxim Mkrtchyan were involved in data acquisition and analysis and worked on aspects of the experiment. All authors have read and agreed to the published version of the manuscript.

Conflict of Interest

The authors declare no conflict of interest.

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