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*Представлені результати експериментального дослідження флуктуацій параметрів газового середовища при займанні в моделюючої камері. Запропоновано нетрадиційний підхід щодо дослідження динаміки небезпечних параметрів середовища на основі поточної віконної оцінки лагових кореляцій Пірсона. Встановлено, що стійкою ознакою раннього займання є значне збільшення кореляції нестационарних флуктуацій температури, концентрації чадного газу та диму в газовому середовищі*

*Ключові слова: пожежа, раннє займання, газове середовище, моделююча камера, кореляції флуктуацій, віконна оцінка*

*Представлены результаты экспериментального исследования флуктуаций параметров газовой среды при возгорании в моделирующей камере. Предложен нетрадиционный подход к исследованию динамики опасных параметров среды на основе текущей оконной оценке лаговых корреляций Пирсона. Установлено, что устойчивым признаком раннего возгорания является значительное увеличение корреляции нестационарных флуктуаций температуры, концентрации угарного газа и дыма в газовой среде*

*Ключевые слова: пожар, раннее возгорание, газовая среда, моделирующая камера, корреляции флуктуаций, оконная оценка*

# EXPERIMENTAL STUDY OF THE FLUCTUATIONS OF GAS MEDIUM PARAMETERS AS EARLY SIGNS OF FIRE

**B. Pospelov**

Doctor of Technical Sciences, Professor\*  
E-mail: pospelov@nuczu.edu.ua

**V. Andronov**

Doctor of Technical Sciences, Professor\*  
E-mail: andronov@nuczu.edu.ua

**E. Rybka**

PhD\*

E-mail: rybka@nuczu.edu.ua

**V. Popov**

Doctor of Technical Sciences, Associate Professor\*  
E-mail: popov@nuczu.edu.ua

**A. Romin**

Doctor of Science in Public Administration,  
Associate Professor\*\*  
E-mail: semkiv@nuczu.edu.ua

\*Research Center\*\*

\*\*National University of Civil Protection of Ukraine  
Chernyshevska str., 94, Kharkiv, Ukraine, 61023

## 1. Introduction

Fires are known to cause enormous damage to nature and society. People die and are injured in fires, material and spiritual artefacts are destroyed, deteriorating the quality of habitat, etc. In this case, the source of any fire is the ignition of combustible material. Global statistics indicates that most fires happen in premises (over 39 %) and deaths from fires in premises account for over 80 %. That is why fire prevention in premises is one of the main and priority problems of any state.

To prevent fires in premises, first of all, it is necessary to reliably detect ignition of combustible materials at an early stage before the moment when fire becomes unmanageable. Difficulties of such detection include minor changes of data on dangerous factors of environment, registered by fire sensors. In this case, slight changes of data on hazardous factors of environment are of non-stationary nature and are usually disguised by random perturbations in gaseous medium.

That is why the study of dynamics of the main hazardous factors of gaseous medium in premises at early ignition of the typical combustible materials is relevant.

## 2. Literature review and problem statement

Experimental study of the process of fire occurrence for various premises and materials is considered in papers

[1–3]. It is noted that the process of fire development in the premises has essentially indeterminate and non-stationary nature, depending on many parameters of both the premises themselves and fire load location in it. It should be noted that articles [4, 5] and other known studies explore only dynamics of the basic parameters of gaseous medium mostly in the form of temperature and concentration of combustion products of various materials. In this case, due to complex and non-stationary dynamics of parameters of gaseous medium, other characteristics are not considered.

Paper [6] addresses the enhancement of effectiveness of fire sensors for determining of temperature of gaseous medium in premises during a fire under actual conditions. The problems of self-adjusting fire sensors are discussed in [7, 8]. In this case, articles [6–8] take into account only the unpredictable and non-stationary character of dynamics of the corresponding parameters of gaseous medium. Other characteristics of dynamics that are important for early fire detection, such as correlation of fluctuations of dynamics, are not considered.

Dynamics of carbon monoxide (CO) has been recently used for early ignition detection in premises. That is why fire sensors of CO are most often included in various types of multi-sensor fire detectors as one of the channels for obtaining information about ignitions [9–11]. An important role in early ignition detection is played by dynamics of smoke concentration in gaseous medium of environment during a

fire [12]. However, its use is associated with a high frequency of false detection.

In [13], cross-correlation of dynamics of CO and smoke concentrations during fire experiments in premises is explored. It is shown that the use of the specified cross-correlation allows us to detect reliably ignitions for test fire centers EN 54 [14, 15]. Study [16] is devoted to experimental research into application of such cross-correlation in the presence of interferences. In research [16], it is shown that by combining data on dynamics of CO and smoke concentration, it is possible to detect most ignition centers in the face of interference. The obtained results are based only on the study of cross-correlation dynamics of CO and smoke concentration. Important correlations of fluctuations of dynamics of CO and smoke concentration and other parameters of gaseous medium are not considered.

An important area of premises in terms of fire detection is the ceiling jet zone, in which characteristic non-stationary changes in parameters of gaseous environment occur [17]. Paper [18] contains results of research into dynamics, auto-correlations and paired correlations of parameters of gaseous medium in the ceiling zone, simulating the chambers at ignition of alcohol, paper, timber and textiles. It was shown that a sign of early ignition detection can be a degree of paired correlation of fluctuations of parameters of gaseous medium with zero lag. It is noted that presence of ignitions on the assigned interval of analysis causes a slight increase in time constant of fluctuations of the studied parameters in gaseous media compared with the interval of absence of ignition. However, this approach has a low sensitivity for identifying ignitions at an early stage. In this case, correlations of fluctuations of parameters of gaseous medium that are important for the early ignition detection are not considered.

Thus, literature analysis shows that nowadays there are no studies of correlation of fluctuation of dynamics of main gaseous medium parameters at an early ignition of materials in the premises. Such studies are essential for development of new technology for reliable detection of an ignition of materials in the premises. That is why experimental study of the correlation of fluctuations of dynamics of basic gaseous medium parameters as non-stationary processes is an important and unresolved part of the problem of fire detection at the early stages.

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### 3. The aim and objectives of the study

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The aim of present research is to study correlations of fluctuations of dynamics of basic parameters of gaseous medium at an early ignition of materials based on experimental data, obtained at ignition of typical materials in a simulation chamber.

To accomplish the set goal, the following tasks were to be solved:

- to state conditions of the experimental research into correlations of fluctuations of dynamics of basic parameters of gaseous medium at an early ignition of material in the chamber;
- to substantiate the theoretical base of the experimental study;
- to explore correlations of fluctuations of dynamics of the basic parameters of gaseous medium at an early ignition of alcohol, paper, timber and textiles.

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### 4. Conditions of the experimental study of fluctuations correlations of dynamics of gaseous medium parameters

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Premises are usually characterized by an assigned fire load, determined by the quantity of combustible materials and properties. Dynamics of hazardous factors of ignition of materials in premises depends not only on fire load, but also on ventilation of premises. A hotbed of early ignition is traditionally regarded as a source of perturbations of the basic parameters of gaseous medium (ambient temperature, concentrations of smoke and CO). In this regard, early ignition detection in premises can be carried out based on recording dynamics of specified parameters of the environment in real time or analysis of fluctuations of the specified dynamics. Fluctuations of dynamics typically characterize temporary instability of parameters of gaseous medium in real-time scale, which can be used to identify the ignition moment.

This necessitates experimental study of correlation of fluctuation of dynamics of basic gaseous medium parameters at an early ignition of alcohol, paper, timber and textiles in a simulating chamber in the actual scale of monitoring time.

Experimental studies were carried out in a special chamber [19], imitating pressurized premises, at ignition and combustion of test combustible materials, such as alcohol, paper, timber and textiles. The volume of the chamber made up 0.524 m<sup>3</sup>, in the upper part of the chamber at the height of 0.84 m, there were sensors that registered physical parameters of gaseous medium at ignition of correspondent combustible materials. In this case, the height of the flame for alcohol, paper and timber accounted for less than 0.2 m. Data  $x_n$  on parameters of gaseous medium at ignition of the specified materials were registered by sensor of temperature (detector TPT-4), optical smoke sensor (detector SPD-3.2) and carbon oxide sensor (of Discovery series). Data registration was carried out at discrete moments  $t_n$ , where  $n=1, 2, \dots, 3,000$ . Ambient temperature, concentration of CO and smoke were registered as parameters of gaseous medium in the chamber at ignition. The data, measured by the sensors, were recorded to random access memory of computer. Special software allowed reading the relevant sensors with different frequency. In this experiment, reading of sensors was carried out with frequency of 10 Hz (time interval of 0.1 s). Ignition of materials was conducted approximately at moment  $t_{200}$ , corresponding to 20 s.

Because the initial stage of ignition was of a special interest, the time interval that a little exceeded ignition moment  $t_{200}$  was selected as the interval of analysis. In this case, this interval was 30 s. Registration of data on parameters of medium in the chamber and estimation of correlations of fluctuations of the specified parameters were performed at the pace of data arrival within the explored interval of analysis. Estimation of correlations of fluctuations of gaseous medium parameters in the chamber was carried out based on calculation of rolling (with the assigned window) of autocorrelation function, used for correlation analysis of non-stationary processes. The study of correlation of fluctuations of gaseous medium parameters at an early ignition was conducted for each of the indicated combustible materials in the following sequence: alcohol, paper, timber and textiles. After each session of ignition analysis there was natural ventilation of the chamber for 5–7 minutes for bringing parameters of gaseous medium in the chamber to the state that was close to the initial.

**5. Theory behind the experiment**

Gaseous medium in the chamber in the absence of ignition is a system in equilibrium. Let us characterize parameters of such a system in equilibrium by certain values of temperature, concentration of smoke and CO. Experimental study of dynamics of specified parameters of gaseous medium at various states of equilibrium before ignition of alcohol, paper, timber and textiles in the chamber, was considered in [20]. It was established that emergence of ignition in the chamber disrupts equilibrium of the specified parameters of the medium and leads to an increase in the autocorrelation between different counts of each parameter. In this case, autocorrelation of the considered parameters of the medium was studied, assuming ergodicity of parameters of the medium on the limited intervals of analysis of ignition absence and presence. This means that under actual conditions, ignition can be detected only after registration of all data on the analysis interval. In [20], intervals of analysis make up 20 s. Enhancement of accuracy of this correlation analysis by increasing the observation interval leads to narrowing down the differences in correlation parameters of the medium in the absence and presence of ignition. This is explained by the incorrect use of the methods of stationary correlation analysis in the explored non-stationary case.

In contrast to [20], we will assume that parameters of gaseous medium in the chamber on fairly lengthy observation intervals at ignition of alcohol, paper, timber and textiles are non-stationary processes. In this case, the right boundary of the observation interval is not fixed, but rather determined by the current moment of observation. These assumptions are closer to actual conditions of early detection in comparison with the ergodic interval approach.

In this case, first it is proposed to transform the observed non-stationary parameters of gaseous medium into current increments  $y_j$  of the corresponding parameter, determined by  $y_j = x_{j+1} - x_j$  for  $j = 0, 1, \dots, 300$ . Because values of parameters  $x_{j+1}$  and  $x_j$  differ by one discretion step, it can be argued that their mean values will be slightly different. This means that magnitude  $y_j = x_{j+1} - x_j$  will determine increment of the correspondent parameter, caused by its non-stationary dynamics. By virtue of random character, dynamics of increment of the corresponding parameter will determine its non-stationary fluctuations, caused by instability of equilibrium of parameters of the medium.

To study correlations of non-stationary fluctuations of parameters of the medium, we will use the Pearson lag correlation coefficient, calculated for a fixed-size rolling window  $w$ . Taking this into consideration, the Pearson window lag correlation coefficient for the current moment of time  $t=j$  will be determined in accordance with the expression

$$RP_y(t, \tau, w) = \text{if} \{t < \tau, r_y(t, \tau), r_y(t, \tau, w)\}. \tag{1}$$

In this case, in expression (1) function

$$r_y(j, \tau) = \frac{\sum_{i=0}^j y_i y_{i+|\tau|}}{\sqrt{\sum_{i=0}^j y_i y_i \cdot \sum_{i=0}^j y_{i+|\tau|} y_{i+|\tau|}}}$$

and function

$$r_y(j, \tau, w) = \frac{\sum_{i=j-w+1}^j y_i y_{i+|\tau|}}{\sqrt{\sum_{i=j-w+1}^j y_i y_i \cdot \sum_{i=j-w+1}^j y_{i+|\tau|} y_{i+|\tau|}}}$$

where  $\tau$  is the magnitude of lag. It should be noted that current correlation coefficient (1) is a window lag coefficient and has the best interpretability of calculation results. However, this coefficient is best suited for evaluation of a linear relationship between two arbitrary values of the process with Gaussian distribution. In the case of non-Gaussian distribution of the values of the explored process, it will continue to characterize a degree of linear relationship of the studied values of the analyzed process, but it will be impossible to apply methods of verification for significance to it.

Given the absence of a strict theory of values of correlation coefficient, we will adhere to the following levels: from 0 to 0.2 – very weak; from 0.2 to 0.5 – weak; from 0.5 to 0.7 – average; from 0.7 to 0.9 – high; from 0.9 to 1 – very high. In accordance with the above theoretical information, experimental studies of correlation of fluctuations of the main parameters of gaseous medium in the chamber were conducted.

**6. Results of the experiment to study correlations of fluctuations of dynamics of gaseous medium parameters**

Research into correlation of non-stationary fluctuations of the main parameters of gaseous medium was carried out based of the current Pearson lag correlation coefficient with rolling window  $w=20$ . In this case, results of the research into current correlation coefficient of fluctuations of the medium to a very high level.

Fig. 1 shows the results of the study of current correlation coefficient of fluctuations of temperature in the chamber (in coordinates of discrete countdown  $t$  and lag number  $\tau$ ) at early ignition of alcohol, paper, timber and textiles. In this case, the corresponding area for the conditional level of current value of lag correlation coefficient is displayed in color in the Figures. The minimum level is displayed in blue, and the maximum level is displayed in red.

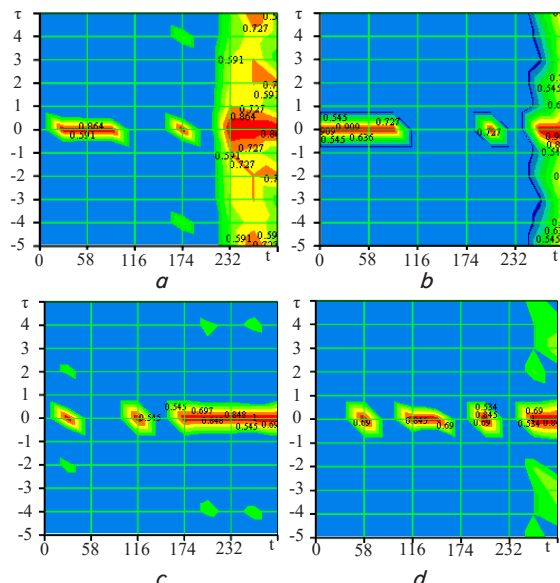


Fig. 1. Current lag correlation coefficient of temperature fluctuations of gaseous medium in the chamber: a – alcohol; b – paper; c – timber; d – textiles

In the course of the study, there was natural ventilation for 5–7 minutes of the gaseous medium in chamber between ignitions of materials.

Similar results of the study of current lag correlation coefficient of fluctuations of carbon monoxide concentration are shown in Fig. 2, and of CO concentration are shown in Fig. 3.

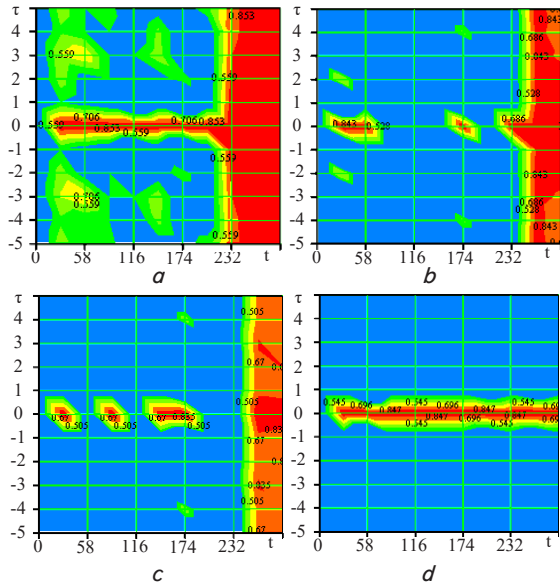


Fig. 2. Current lag correlation coefficient of fluctuations of carbon monoxide concentration in gaseous medium of the chamber: *a* – alcohol; *b* – paper; *c* – timber; *d* – textiles

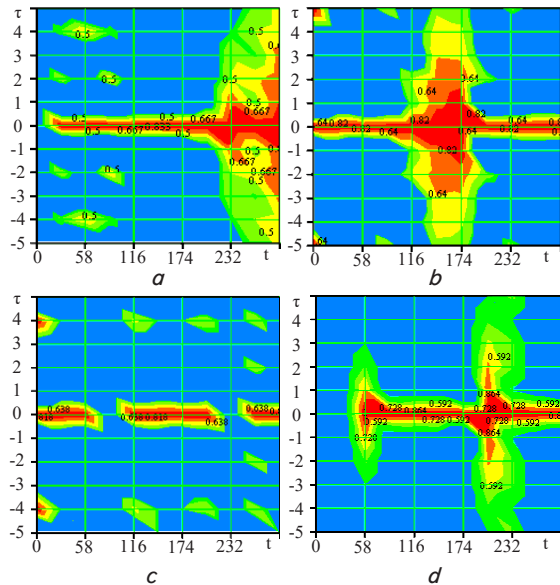


Fig. 3. Current lag correlation coefficient of smoke concentration fluctuations in gas medium of the chamber: *a* – alcohol; *b* – paper; *c* – timber; *d* – textiles

Research results, shown in Fig. 1–3, were obtained, taking into account errors of measurements of parameters in gaseous medium by the sensors in the chamber. Errors of data registration during their transformation into a digital form were taken into account. In this case, errors of data transformation into a digital form can be considered negligible compared with errors of sensor measurement. In this regard, obtained results can be used for comparative estimation of current correlations of fluctuations of the corresponding parameters of gaseous medium in the chamber at an early ignition of test combustible materials taking into account

sensor errors. It should be noted that the sensors, used in the experiment, are applied in actual fire detectors. That is why it can be argued that the studied fluctuations of parameters of gaseous medium are caused by emergence of a source of early ignition in the chamber and errors of correspondent measuring sensors. This means adequacy of the conditions of the experiment to actual conditions.

**7. Discussion of results of research into correlations of fluctuations of dynamics of gas medium parameters**

Based on analysis of the data in Fig. 1–3, it can be assumed that ignition of combustible material that occurs violates the equilibrium of parameters of gaseous medium in the chamber and causes diffusion of parameters of the medium by the laws that differ from the known ones. In this case, fluctuations of parameters of gaseous medium at different stages can be described by random processes with varying increment correlation degrees. In the absence of ignitions, they are described by autoregression processes of the order from 1 to 10. For most of the studied parameters, these are processes of the order from 1 to 3. It should be noted that based on results of research into current lag correlation coefficient of fluctuations, shown in Fig. 1–3, it is possible to determine a more accurate order for correspondent autoregression processes. In this case, emergence of ignition in the chamber leads in the majority of cases to expansion of correlation relationship to neighboring fluctuation of parameters of gaseous medium and a corresponding increase in the order of autoregression processes.

Calculation of current lag correlation coefficient of fluctuations of parameters of gaseous medium in the chamber makes it possible, in contrast to [20], to localize the time of emergence of early ignition of combustible materials. In addition, it was established that temporary localization of the moment of early ignition is accompanied by expansion of the correlation relationship between neighboring fluctuations of the corresponding parameters of gaseous medium.

Thus, for example, analysis of the data in Fig. 1 shows that fluctuations of ambient temperature in the chamber enables us in real-time monitoring of this parameter to localize the moment of starting of ignition for alcohol and paper. Somewhat worse possibilities were observed at early ignition of textiles and timber in the chamber. This is explained by the low rate of ignition of these materials. In this case, subsequent temperature monitoring allows localization of an early ignition of these combustible materials. The time of localization in this case somewhat exceeds the observation interval, specified in the experiment.

Analysis of the data in Fig. 2 illustrates the fact that fluctuations of carbon monoxide concentration in the medium of the chamber make it possible in real-time measurement of this parameter to localize the moment of beginning of ignition for alcohol, paper and timber. At early ignition of textiles in the chamber, the possibilities are a bit worse. This is explained by special features of textiles ignition, associated with a longer stage of pyrolysis and smoke release. In the course of subsequent monitoring of CO concentration, it is possible to localize in time the moment of textiles ignition. However, the localization time in this case also exceeds the interval of analysis, assigned in the experiment.

Data in Fig. 3 show that fluctuations in smoke concentration in gaseous medium of the chamber enables us in

real-time parameter measurement to localize the moment of beginning of ignition for alcohol, paper and textiles. At early ignition of timber in the chamber, the possibilities are a bit worse. This is explained by the peculiarities of timber ignition, associated with a small amount of smoke release at an early stage of ignition. In the course of subsequent monitoring of smoke concentration, it is possible to localize in time the moment of timber ignition. However, the localization time in this case exceeds the interval of analysis, assigned in the experiment. Obtaining of original results can be considered the merit of this study. These results are an attempt of experimental identification of special features of early ignition of typical combustible materials in pressurized premises, which is based on analysis of current correlations of fluctuations of parameters of gaseous medium in the chamber. Obtained results can also be used for early detection of ignitions. The shortcomings of the study can include restrictions, imposed by dimensions of the test chamber, location of measurement sensors and conditions of insufficient chamber ventilation after ignition of each sample. It is advisable to continue subsequent research based of the proposed approach towards considering actual pressurized premises with different fire load.

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## 8. Conclusions

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1. An unconventional approach to research into dynamics of the main hazardous factors of gaseous medium in premises at early ignition of combustible materials was proposed. This approach is based on current window estimation of the Pearson lag correlations of fluctuations of dynamics of gaseous medium parameters as non-stationary processes.

In contrast to known approaches, a given approach makes it possible to perform reliable localization of early ignitions of combustible materials in premises.

2. It was experimentally determined that early ignition of combustible materials has a significant impact on correlations of fluctuations of the measured parameters of gaseous medium, such as temperature and concentrations of carbon monoxide and smoke. Correlations of fluctuations of carbon monoxide and smoke concentrations are most informative in terms of localization of early ignitions of the considered combustible materials. Temperature fluctuations are more informative at localization of early ignition of alcohol and paper.

3. Results of current window estimation of the Pearson lag correlations for fluctuations of dynamics of the basic gaseous media parameters were obtained. It was shown that in the absence of ignition, fluctuations can be considered non-correlated with sufficient accuracy for practical applications. It was established that the presence of ignition leads to occurrence of non-stationary correlations of fluctuations. In this case, fluctuations can be described by higher-order autoregression processes, determined by the kind of combustible material and the stage of its ignition development. It has been established that a sustainable sign of early ignition of combustible materials in the simulation chamber is a significant increase in the interval of correlation of non-stationary temperature fluctuations, as well as concentrations of carbon monoxide and smoke in gaseous medium of the chamber. That is, information about the beginning of early ignition lies in the low-frequency region of non-stationary spectrum of fluctuations of the corresponding parameter of gaseous medium. In equilibrium, fluctuations have much smaller correlation interval, characteristic for uncorrelated processes.

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*Досліджено процеси електрохімічного видалення іонів кадмію та цинку окремо і в суміші з розчинів сірчаної або соляної кислот в одно- та двокамерних електролізерах. Показано графічну залежність виходу за струмом важких металів від початкових характеристик розчинів та умов процесу. Доведена перспективність використання електролізу для селективного видалення важких металів із суміші з кислих розчинів*

*Ключові слова: іонний обмін, електрохімічні методи очищення води, стічні води, відпрацьований регенераційний розчин*

*Исследованы процессы электрохимического удаления ионов кадмия и цинка отдельно и в смеси из растворов серной или соляной кислот в одно- и двухкамерных электролизерах. Показана графическая зависимость выхода по току тяжелых металлов от начальных характеристик растворов и условий процесса. Доказана перспективность использования электролиза для селективного удаления тяжелых металлов из смеси из кислых растворов*

*Ключевые слова: ионный обмен, электрохимические способы очистки воды, сточные воды, отработанный регенерационный раствор*

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# ELECTROEXTRACTION OF HEAVY METALS FROM WASTEWATER FOR THE PROTECTION OF NATURAL WATER BODIES FROM POLLUTION

**M. Gomelya**

Doctor of Technical Sciences,  
Professor, Head of Department\*

E-mail: m.gomelya@kpi.ua

**G. Trohymenko**

PhD, Associate Professor

Department of Environmental and Labor Safety  
National University of Shipbuilding

Heroiv Stalinhrada ave., 9, Mykolaiv, Ukraine, 54025

E-mail: antr@ukr.net,

**O. Hlushko**

PhD, Associate Professor\*

E-mail: alyona\_glushko@ukr.net

**T. Shabliy**

Doctor of Technical Sciences, Associate Professor\*

E-mail: dsts1@ukr.net

\*Department of Ecology and Technology of Plant Polymers  
National Technical University of Ukraine  
“Igor Sikorsky Kyiv Polytechnic Institute”  
Peremohy ave., 37, Kyiv, Ukraine, 03056

## 1. Introduction

In the classic sense, the class of “heavy metals” includes zinc, chromium, copper, cadmium, cobalt, nickel, arsenic, selenium, silver, antimony, mercury, thallium and lead. Com-

pounds of these elements as well as the elements themselves are toxic [1]. About 70 % of toxic metals get into a human body with food. The most dangerous toxic elements in foods to be controlled are mercury, cadmium, lead, arsenic, copper, tin, zinc, and iron. High probability of population poisoning