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SOFTWARE FOR THE CERTIFICATION OF IODINE-IMPREGNATED SORBENTS OF AIR FILTERS OF NPP

M.F. Kozhevnikova
National Science Center "Kharkiv Institute of Physics and Technology", Kharkiv, Ukraine
E-mail: mar-fik@ukr.net

Ukraine has a developed network of nuclear power plants (NPP) that produce a significant part of the total electricity. The issues of safety and reliability of nuclear power are the most important both from the point of view of ensuring the quality of life of the population and ensuring the energy needs of the economy as a whole. To purify atmospheric air at nuclear power plants, carbon filters with sorbent in the form of granules with a branched pore system are used, which are able to capture and retain iodine and methyl iodide molecules. An important characteristic of sorbent is the degree of purification of gases passed through them and the permissible service life of filters without replacing granules. The purpose of this work was to create software for automatically determining the filter purification coefficient based on processing the measured spectra of the X-ray of the studied and standard samples of sorbent and printing the certification protocol. As a result of the work, computer code was developed in the Delphi programming language. Software has been created for automatic processing of experimental data on methyl iodide accumulated in the main and additional sections of the air purification filter and calculation of the filter purification coefficient.

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INTRODUCTION

Ukraine has a developed network of nuclear power plants that produce a significant part of the country's total electricity. The part of nuclear power plants in the total energy balance was more than 50% [1] even before the beginning of the Russian aggression and increased even more due to the destruction of the thermal power plant and GRES system [2]. The issues of safety and reliability of nuclear energy are the most important both from the point of view of ensuring the quality of life of the population and ensuring the energy needs of the economy as a whole. The functioning of nuclear power plants requires the control of many mechanisms related to the formation and distribution of radioactive substances. One of the main sources of the penetration of radioisotopes into the environment are radioactive gases, among which radioactive iodine ^{131}I. Gas with ^{131}I can be present in the atmosphere of nuclear power plants in the form of molecular iodine I_2 itself or the compound – methyl iodide, which has the molecular formula CH_3I, and is a particular danger to human health. ^{131}I has a short half-life of 8.04 days, but both iodine-containing gases are very volatile.

^{131}I has an extremely negative effect on the functioning of the thyroid gland, which absorbs about 50% of the iodine entering the body due to the presence of iodine-containing hormones in its composition – thyroxine and triiodothyroain. Due to the absorption of radioactive iodine by the body, its atoms are replaced by stable ones, which lead to dysfunction of the thyroid gland and the risk of malignant tissue degeneration. Iodine-131 enters the human body mainly through the lungs, which makes it necessary to thoroughly clean the atmospheric air of the nuclear power plant from radioactive iodine-containing gases.

1. THE TASK OF WORK

The carbon filters with sorbents in the form of granules with a branched pores system capable of capturing and retaining iodine and methyl iodide molecules are used to clean atmospheric air at nuclear power plants. An extremely important characteristic of sorbents is the degree of purification of the gases passing through them and the permissible period of operation of the filters without replacing the granules. Each type of sorbent is subject to certification, which is carried out by performing certain procedures: an emergency situation is modeled by pre-saturating the pores of the granules with water molecules by long-term pumping of air saturated with water vapor. On the next stage a vapor-gas mixture containing iodine or methyl iodide is passed through the filters. The percentage of iodine-containing gases retained in the filter substance is determined after the end of pumping. A satisfactory indicator is 99 or more percent.

There are two main methods of determining iodine/methyl iodide accumulated in the substance of sorbents during tests. The first one can be implemented if radioactive iodine is used in the certification of filters – isotope ^{131}I, or methyl iodide with a radioactive iodine isotope in the molecule [3]. In this case, iodine deposited in the filter is determined by gamma emission of quanta with energies of 634 and 364 keV, accompanied by γ-decay of the nucleus of the isotope ^{131}I atom into the nucleus of the ^{131}Xe atom. In this case, it is important to conduct measurements quickly enough, since the activity of radioactive iodine decreases by half every 8 days, and if the activity in the final part of the filter was initially small, problems with the accuracy of measurements may arise. This method requires the organization of working conditions with toxic and radioactive substances, the presence of radioactive iodine/methyl iodide and is accompanied by all the complexities inherent in such technologies – the organization of dose control and personnel protection, the disposal of toxic and radioactive waste, the presence of spectrometric equipment, software for data processing and preparation of the test report.

The second method involves the use of iodine and methyl iodide with stable isotopes of iodine in the composition of the molecules. In this case, the organization of research is significantly simplified, since there is no radioactive emission, and the analysis of the iodine content is carried out by means of recording the X-ray of iodine K-series excited by accelerated protons, by X-ray emission from a tube [4] or γ-emission from a radioactive source [4-7]. Spectrometric equipment and software for primary processing of emission spectra is similar to that used when measuring activity when using ^{131}I isotope.

The second method looks safer for personnel and the environment. But during the certification of certain brands of sorbent, difficulties arise due to the initial presence of iodine in the sorbent granules. We are talking about the category of iodine-impregnated sorbent in which the iodine content can vary from 0 to 1.5…2.5% in individual granules. It is assumed that the presence of stable iodine in sorbents makes an additional contribution to the retention of radioisotopes, since, unlike ordinary iodine, the main absorption mechanism of which is physical adsorption, in the case of ^{131}I, the retention efficiency is determined by both the porous structure of the sorbent used and the chemisorption component, which depends on the presence of impregnants. The use of iodine-impregnated sorbents and the recorded significant variation in the concentration of iodine in the sorbent granules of the same series leads to the fact that at the certifying similar materials, it is necessary to perform two series of measurements, the first – before pumping methyl iodide, to determine the initial content of iodine in filters, second – after testing, to determine the amount of iodine/methyl iodide retained by the filters. Determination of iodine content is carried out by the method of an external standard. The content of iodine in each of the six sections of the filter before and after pumping, as well as in two control additional sections after the filter, is analyzed separately. The filter cleaning coefficient K is determined by the formula:

$$K = \frac{M_F^I}{M_F^I + M_D^I}\ , \tag{1}$$

where M_F^I is the total mass of iodine/methyl iodide deposited in main sections of the filter during the pumping M_D^I the total mass of iodine/methyl iodide deposited in additional sections of the filter during the pumping.

Thus, the content of iodine is determined in six main and two additional sections before and after pumping. For each section, three parallel measurements of X-ray of iodine K- series spectra are made and also two series of parallel measurements of the spectrum of a standard comparison sample with a known iodine content.

The sum of the counts under the peaks of the analytical lines of iodine in the X-spectra is determined. The mass of iodine deposit as a result of pumping in the main and additional filter section is calculated on the basis of the processing the spectrometric information. The filter cleaning coefficient is determined by formula (1) as result of measurements and processing of X-ray spectra.

The purpose of this work was the creation of software for the automatic determination of the filter cleaning coefficient based on the processing of the measured x-ray spectra, test and standard samples, before issuing the certification protocol for printing.

2. ANALYSIS METHODS

A computer code for the automatic processing of experimentally measured X-ray spectra of samples of air filter NPP was developed. The filter cleaning coefficient calculates based on the quantities of methyl iodide determined in the main and additional sections of filter.

The software is developed in Delphi programming codes. Delphi is a structured, object-oriented, high-level programming language with strict static variable typing. The main area of use is writing application software [8].

3. RESULTS OF THE RESEARCH

In the course of work, the "Filter" program was created to calculate the coefficient of atmospheric air purification by NPP coal filters during tests at the SKIF stand. This program was developed with the aim of automating the procedure for processing the results of measurements at the SKIF stand, created at IFTTMT [9].

The Filter program is intended for processing X-ray spectra: viewing and calculating peak areas for one spectrum and a group of spectra. And also for calculating the integral coefficient of retention of methyl iodide by a multi-section filter filled with coal sorbent granules used in the air purification system of the NPP.

To determine the quantitative content of methyl iodide accumulated in the sorbent substance, the X-ray spectral analysis (XSA) method is used [7]. Gamma radiation from a ^{241}Am source with an energy of 59.54 keV is used for fluorescent excitation of the characteristic X-ray radiation of the K-series of iodine with an energy of 28.18 keV (K-edge absorption of 30 keV). A series of parallel measurements with an external standard is made to determine the mass of iodine in sorbent samples selected from ten basic and two control sections of the filter, before and after pumping.

The software was developed in three stages:

1. Creation of an algorithm for the software implementation of the task "Calculation of the coefficient of atmospheric air purification by carbon filters of nuclear power plants during tests at the SKIF stand".
2. Programming in the Delphi7 environment. Step-by-step development of software modules.
3. Seal of the certification protocol.

The Filter program consists of 5 modules to perform the task:

1) The FILE module (Fig. 1) is used to read data and record processing results.The following features are provided in this module:

– download of the spectrum (with visualization on the screen);

– loading spectrum groups;
– recording of processing results for the spectrum;
– recording of the task file after processing the spectrum;
– recording of results for a group of spectra;
– download spectrum groups for iodine.

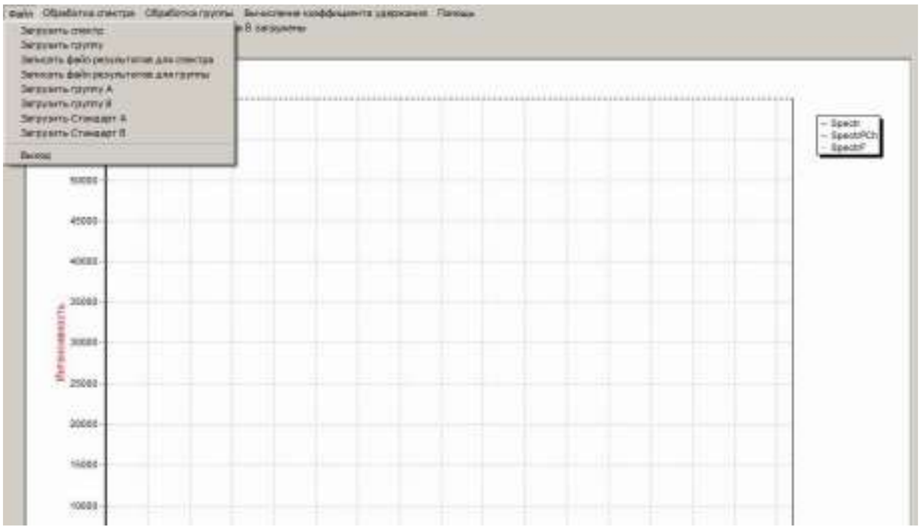

Fig. 1. Working window of the Filter program: FILE module

2) The SPECTRUM PROCESSING module (Fig. 2) performs the task of obtaining the background in the selected part of the spectrum (the Chebyshev orthogonal polynomial method is used and calculating the peak area in the selected interval.

The following features are provided in this module:
– input of text windows for peak intervals, degree of polynomial, peak number;
– creating an information window that displays information after processing the spectrum: degree of the polynomial, root mean square deviation at a given degree of the polynomial, peak area and error;
– background adjustment of the spectrum at the given interval.

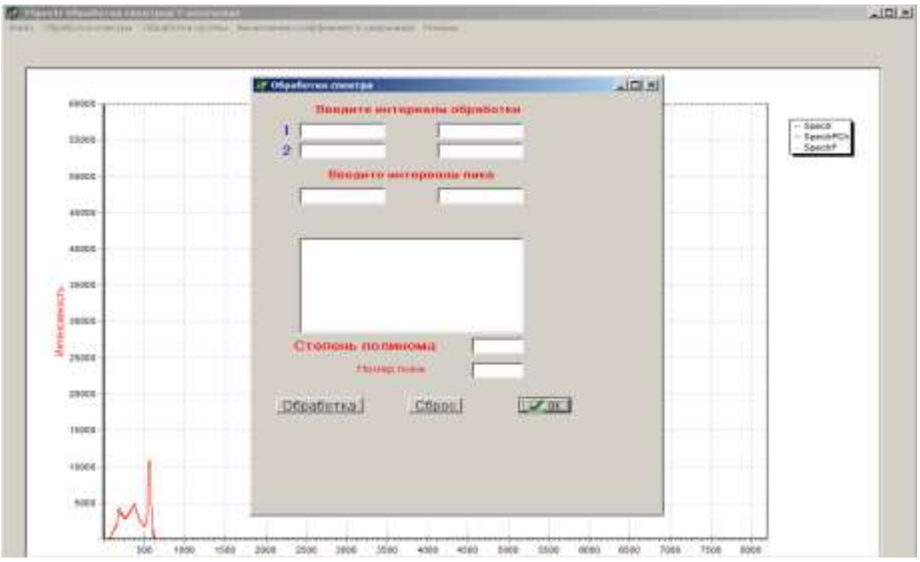

Fig. 2. Working window of the Filter program: SPECTRUM PROCESSING module

3) The GROUP PROCESSING module (Fig. 3) performs the task of calculating the peak area at selected intervals for a group of similar spectra.

The following features are provided in this module:
– download the assignment file for the spectrum group using the FILE ASSIGNMENT button;
– calculation of peak areas and errors, output of the information window with data calculated for each spectrum (PROCESS button).

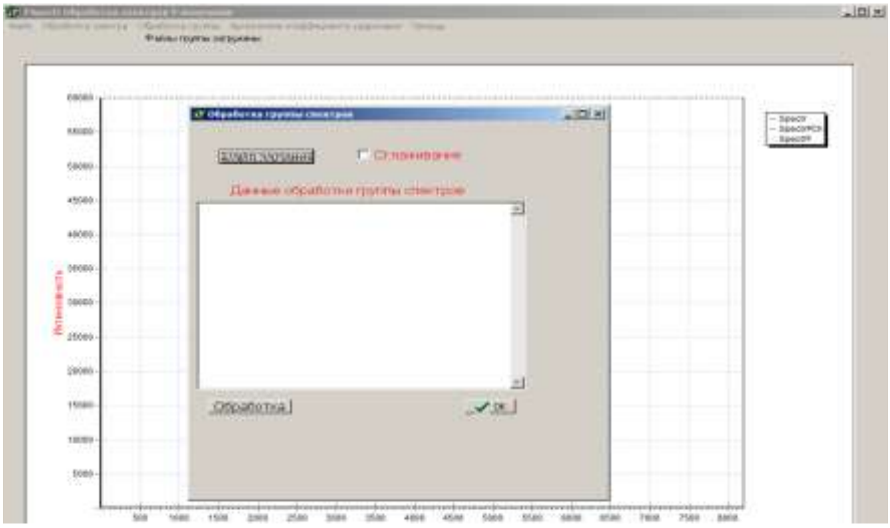

Fig. 3. Working window of the Filter program: GROUP PROCESSING module

4) The DETERMINATION OF IODINE MASS module (Fig. 4) performs the task of calculating the coefficient of air purification from CH_3I.

The following features are created in this module:

– entering the task file for the spectrum group;

– entering a file with values of the depth of placement of samples in the filter;

– entering a file with sample mass values;

– text window MASS OF IODINE IN THE STANDARD;

– text box QUANTITY OF MEASURED SAMPLE;

– the output of the obtained results is calculated on the screen;

– recording of calculated results in an EXCEL file.

The following are provided for the studied group of spectra:

– calculation of the sum of the SUM peaks for the iodine Kα-series line in the spectra of the sorbent samples before and after pumping, as well as in the spectra of the standard sample;

– calculation of the average value of the ratio of the mass of iodine in the standard sample to the sum of the counts in the analytical peak K=M/SUM for the spectra of the standard. Separately – for measurements before pumping, separately - after pumping;

– calculation of the mass of iodine C for each sample before pumping, the results of n parallel measurements are averaged;

– calculation of the mass of iodine M for each spectrum of each sample after pumping. For each sample, n results of parallel measurements are obtained. A total of N*n concentration values divided into N groups of n values;

- calculation of the mass of iodine added as a result of pumping;

– construction of a table of the dependence of the added iodine content on the thickness of the sorbent layer;

– seal of the certification protocol.

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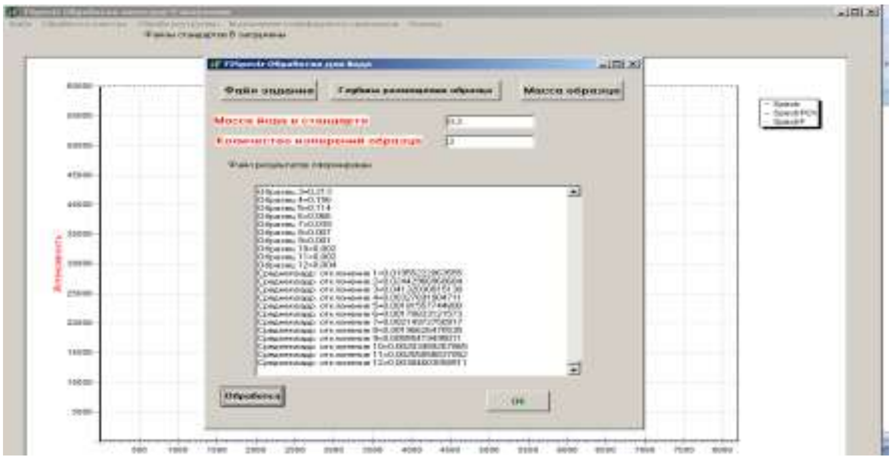

Fig. 4. Working window of the Filter program: module DETERMINATION OF IODINE MASS

The HELP module contains instructions for using the program.

The Filter program for calculating the coefficient of atmospheric air purification by NPP coal filters during tests at the SKIF stand provides for automated printing of the certification protocol (Fig. 5).

Протокол испытаний №		
Лабораторные испытания по определению показателя эффективности удержания аэрозольных эффективных йодистых соединений сорбционными материалами		
Согласно процедуре контроля качества №____ организации по надзору		
ННЦ ХФТИ в соответствии со стандартом безопасности №_____		
Происхождение образца:	компания (неиспользованный/использованный активированный уголь)	
Результаты испытаний:	Эффективность удержания %	0,993941944
Примечания:	Показатель эффективности удержания, приведенный в результате испытаний, указан для испытатетельного слоя толщиной 0.5 м	

Результаты испытаний		
Толщина слоя (см)	Время пребывания и удержания (с)	Эффективность удержания (%)
5		0,237406901
10		0,443415961
15		0,639907937
20		0,783476954
25		0,888480333
30		0,949018908
35		0,984901077
40		0,99124644
45		0,992072607
50		0,993941944
55		0,995976208
60		1

Fig. 5. Example of a certification protocol

CONCLUSIONS

This work was carried out within the framework of the targeted comprehensive program of scientific research of the National Academy of Sciences of Ukraine "Problems of resources and safety of operation of structures, buildings and machines".

The development of a computer code in the language of Delphi programming was made for automatic processing of experimentally measured spectra of X-ray of the main and sections additional sections of the air purification filter and standard samples. The filter cleaning coefficient is calculated as a result of data processing.

A certification protocol is printed as a result of data processing.

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ПРОГРАМНЕ ЗАБЕЗПЕЧЕННЯ ПРОЦЕДУРИ СЕРТИФІКАЦІЇ ЙОДИМПЕРЕГНОВАНИХ СОРБЕНТІВ ПОВІТРЯНИХ ФІЛЬТРІВ АЕС

М.Ф. Кожевнікова

Україна має розвинену мережу атомних енергоблоків, які виробляють значну частину загальної електроенергії. Питання безпеки та надійності атомної енергетики є найважливішими як з точки зору забезпечення якості життя населенню, так і забезпечення енергетичних потреб економіки загалом. Для очищення атмосферного повітря на атомних електростанціях використовують вуглецеві фільтри з сорбентами у вигляді гранул з розгалуженою системою пір, які здатні захоплювати та утримувати молекули йоду та метилйодиду. Важливою характеристикою сорбентів є ступінь очищення газів, що пропускаються через них, і допустимі терміни експлуатації фільтрів без заміни гранул. Метою даної роботи було створення програмного забезпечення для автоматичного визначення коефіцієнта очищення фільтра на підставі обробки виміряних спектрів х.р.в. досліджуваних та стандартних зразків та виводу на друк протоколу сертифікації. У результаті роботи розроблено комп'ютерний код мовою програмування Delphi. Створено програмне забезпечення для автоматичної обробки експериментальних даних щодо метилйодиду, накопиченого в основних та додаткових секціях фільтра очищення повітря та обчислення коефіцієнта очищення фільтра.