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IODINE FILTER CONTROL STAND (SKIF) FOR CERTIFICATION CARBON SORBENT VENTILATION SYSTEMS OF NPP

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The article provides information on the creation of the first iodine filter control stand (SKIF) in Ukraine, designed to determine the coefficient of purification of gaseous radioactive emissions from iodine and methyl iodide by sorbents used in ventilation systems at Ukrainian nuclear power plants. The possibility of using a stable isotope of iodine to determine the adsorption capacity of carbon sorbents is substantiated. The development, installation, start-up and adjustment of the pumping module and the measuring module were carried out when using air mixtures with methyl iodide based on the stable isotope of iodine ^{127}I. The developed software allows you to use the stand in automatic mode. The manual for the operation of the SKIF stand was created and the method of determining the coefficient of purification of ventilated air from methyl iodide by carbon sorbents was certified within the framework of the enterprise standard.

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INTRODUCTION

The functioning of industrial nuclear reactors requires the implementation of a set of measures to protect the environment and prevent the negative impact of uranium nuclear decay products on human health. Especially after the accidents in Chernobyl and Fukushima. First of all, we are talking about the control of emissions of radioactive gases into the atmospheric air, among which the radioactive isotope of iodine ^{131}I, which can be a part of molecular iodine – I_2 or methyl iodide – CH_3I, is a significant danger. Filters with sorbents in the form of activated carbon are used to clean the air of these substances at nuclear power plants. The ability of operating filters to absorb emissions with radioactive iodine requires periodic verification, which requires certified devices and methods for determining the content of iodine in the substance of the sorbents. It is known that methyl iodide is characterized by a greater penetrating ability compared to the molecular form of iodine [1], and it was the control of the efficiency of the filters to capture this substance that was the main goal of the proposed development.

The relevance of research in this direction is evidenced by the results of the article [2]: the number of publications in this direction has increased approximately several times.

Methods used in foreign countries for determining the coefficient of purification of ventilated air from molecular iodine and its organic forms by carbon sorbents are based on the use of radionuclide ^{131}I, including in the form of CH_3I which can affect the thyroid gland [3, 4]. The creation of such a methodology in Ukraine requires significant organizational and financial costs: development of a regulatory framework, purchase of special equipment, allocation of special premises, training of specialists with appropriate qualifications, etc.

As part of this work, the SKIF iodine filter control stand was created and tested, which combines the processes of pumping an air mixture containing methyl iodide with a stable isotope of iodine through a mock-up filter with activated carbon, and determining the degree of air purification from this gas by activated carbon.

The urgent need for this method and stand is reflected in the materials of the meeting on this issue of the Ministry of Energy and Coal Industry of Ukraine and the National Nuclear Power Plant of Ukraine from 2013: "Protocol of the meeting on the issue of the lack of methods and technical means for measuring the cleaning coefficient of filters installed on systems at NPPs of Ukraine ventilation" and protocol No. 17 dated 01.28.2020 of the meeting on the issue of meeting the requirements of clause 12.2.17 of the OSPU and the development of "Methods for determining the filter cleaning coefficient during input control, before installation on ventilation systems and during regular inspections".

The purpose of the article is to highlight the results of the work of the National Science Center "Kharkov Institute of Physics and Technology" (NSC KIPT) on the creation of an iodine filter control stand (SKIF) and the development for determining method of the cleaning coefficient adsorbent of carbon filter.

CREATION OF THE SKIF

For a considerable of time, studies of the use of radioactive (^{131}I) and stable (^{127}I) iodine in determining the purification coefficient [5, 6] were carried out at the NSC KIPT, which were also carried out in well-known scientific centers [7–9]. They testified that, when performing actions to determine the purification coefficient with a radioactive substance containing ^{131}I, in comparison with stable iodine ^{127}I, using the processes carried out, including sorption, the obtained result is not affected by the isotopic composition of iodine.

Structurally, the SKIF consists of two autonomous modules – the pumping module (MP), which ensures the passage of the required air flow with an admixture of iodine or methyl iodide through the adsorber layout, and the measurement module (MB) using the X-ray radiometric method of the iodine content in separate parts of the adsorber layout with a sorbent (Fig. 1).

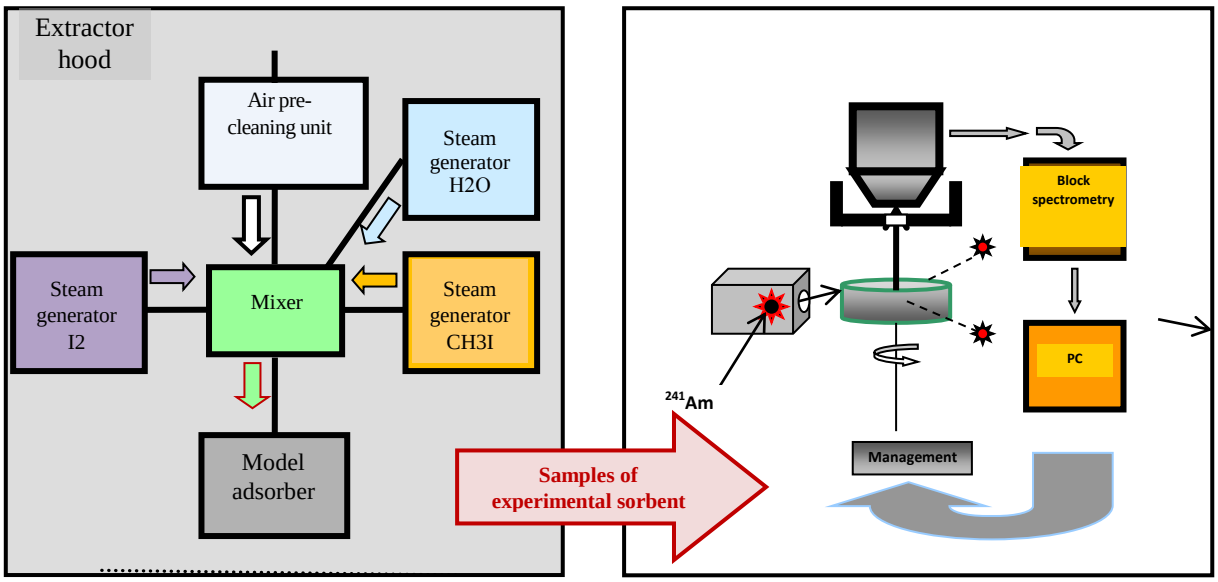

Fig. 1. Composition of modules of the SKIF stand

The data are included in the technical task for the development of the SKIF stand.

When developing the pumping module, we were guided by the following provisions.

1. The mode of operation of the pumping module must correspond to the mode of operation of the AU-1500 filter, which is used at Ukrainian nuclear power plants [10].

2. The module should consist of an iodine pumping unit (pumping air with an admixture of iodine vapors) and a methyl iodide pumping unit (pumping air with an admixture of methyl iodide vapors).

3. The temperature of the sorbent in the adsorber layout should not exceed 55 °C [10].

4. The adsorber layout should consist of sections for the possibility of determining the distribution of adsorbed substances along the height of the adsorbent layer.

5. During the operation of the pumping module, uncontrolled gas impurities from the outside should not enter the adsorber layout.

6. The pumping module should be located in the laboratory fume hood to prevent methyl iodide vapor from entering the operator's respiratory tract when refueling the methyl iodide vapor generator.

For a more confident conclusion about the ability of the method for analyzing the content of iodine in carbon sorbent, the possibilities of various non-destructive nuclear-physical methods of elemental analysis for its determination were investigated at the NSC KIPT: gamma-activation (GAA), X-ray radiometry (RPA), X-ray fluorescence (XRF), characteristic X-ray radiation (PIXE). It is shown that X-ray radiometric analysis most fully meets the defined metrological requirements. In the conducted experiments with RPA, iodine atoms were excited by 59.6 keV radiation from a ^{241}Am radioactive source with the registration of the characteristic K-series X-ray radiation of iodine atoms. The choice of this series is related to its rather high energy (~ 30 keV), which allows the use of the required volume of sorbent material during the analysis.

Experiments conducted on the layout of the installation made it possible to develop the technical requirements for creating a measuring module by the RPA method.

Technical requirements for the measurement module.

A layer with activated carbon is intended for determination of iodine content by X-ray radiometric method. Iodine atoms are excited by radiation from the IHIA-1m-5 (^{241}Am) source. The module must use electronic control of individual nodes based on the operator's commands from the computer. For the compatible functioning of components (stepper motors, photo sensors) and for communication with a personal computer, a controller with a given algorithm of their interaction is required. Registration of analytical radiation is carried out by a detector with the required energy separation. The geometric scheme of sample irradiation and registration of excited radiation is shown in Fig.2.

As a result of the completed design work, installation and start-up, a control stand for SKIF iodine filters was created in the form of two modules: pumping and measurement. The SKIF stand is shown in Figs. 3, 4.

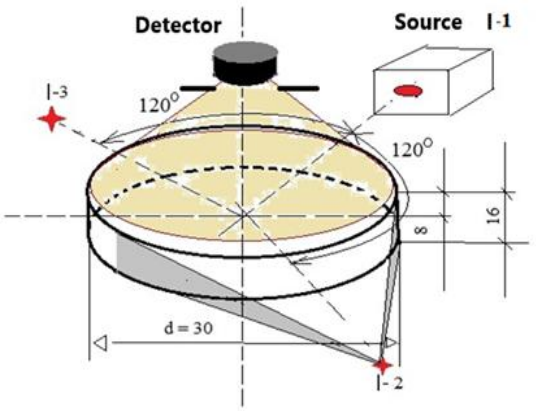

Fig. 2. Geometric diagram of the measuring module

Fig. 3. Pumping module of the SKIF stand

The pumping module is placed in the laboratory cabinet. In the foreground, you can see a yellow gas meter in a mock-up of an adsorber with an electric furnace in a detachable housing located above it. On the panel located in front of the thermostat block, under the inscriptions "H2O", "I2" and "CH3I", valves B1, B2, and B3 are fixed, which are used to regulate the flow of steam of the corresponding substances into the mixing block.

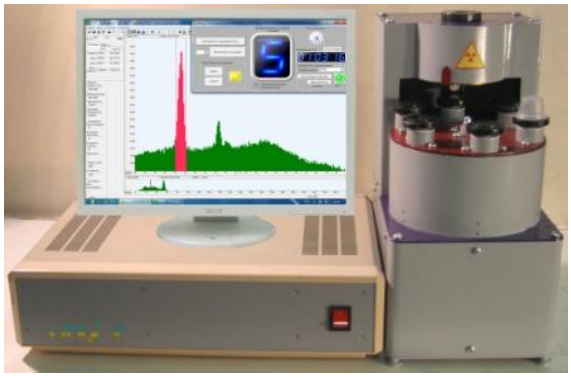

Fig. 4. SKIF stand measurement module (without radiation detector)

The measurement module (see Fig. 4) is designed to determine the mass content of iodine in objects with a carbon matrix (carbon sorbents) by the X-ray radiometric method using the K-series of the characteristic X-ray radiation of iodine, which is excited by modified gamma radiation from a radioactive source with the ^{241}Am isotope. It consists of a control unit, a spectrometry unit and a personal computer.

After the creation of the stand and its testing, the "Instructions for the operation of the SKIF iodine filter control stand" (I 019:2018) was developed and certified by the standardization department of the NSC KIPT, which determined the qualifications of the staff, the scope of application and the conditions of use of the stand [11].

Scope of application of the technique.

The method was developed to determine the coefficient of purification of methyl iodide vapor in the air by carbon sorbents, using non-radioactive materials, and can be applied to the study of carbon sorbents in air filters of ventilation systems at nuclear power plants (NPP). During the operation of the NPP, radioactive fission products leak from the heat-emitting elements into the first circuit of the reactor and then into the working premises of the NPP. The main gaseous radioactive components in the ventilated air are xenon, krypton, iodine vapor and its compounds. Due to its ability to accumulate in the human body, the isotope ^{131}I and methyl iodide, a compound based on it, are particularly dangerous. At present, coal adsorbers of the AU-1500 type, which are designed for filtering ventilated air, are used at NPPs of Ukraine. Reliability of adsorbers ensures radiation safety at NPPs and environmental protection. This technique can be used to determine the content of methyl iodide in various carbon matrices and is suitable for evaluating the effectiveness of the use of carbon filters at NPPs of Ukraine and other nuclear power enterprises, as well as for environmental protection and research. The technique is based on many years of experience in sorption measurements in carbon matrices.

The created stand and methodology allow conducting research on the properties of sorbent samples under conditions that comply with standard D3803 (2022) [3].

In 2022, a comparison was made of the determined iodine absorption coefficient in the laboratory of TÜV SÜD Energietechnik GmbH (Germany) and the NSC KIPT. Coinciding results were obtained.

In addition to this, NSC KIPT received a patent of Ukraine for a utility model No. 153035: "Method of determining the effectiveness of iodine absorption by carbon materials of adsorbers of NPP ventilation systems", which was registered in the State Register in 2023.

EXPERIMENT

The developed stand and methodology were used to conduct a series of experiments aimed at studying factors that can affect the result of determining the activated carbon purification coefficient. Thus, the effect of regeneration of the sorbent at a temperature of 320 °C for 1 h and removal of moisture at 120 °C for 1.5 h was determined. It is shown that the weight of the sorbent changed by 12 wt.% due to the removal of moisture. The saturation of activated carbon with moisture was investigated separately, for which it was aged in atmospheric conditions at a humidity of 35, 50, 75% with an aging period of about 2 weeks.

It was established that long-term storage of the sorbent in non-specific conditions leads to a significant change in the absorption capacity.

CONCLUSIONS

For the first time in Ukraine, the SKIF iodine filter control stand was created, intended for certification of the absorbing properties of adsorbents of NPP ventilation systems.

Based on the SKIF stand, a method for determining the coefficient of purification of ventilated air from methyl iodide by coal adsorbents of NPP ventilation systems has been developed. The methodology regulates the sequence of procedures, taking into account the safety of personnel and the prevention of atmospheric air pollution.

Radioactive iodine isotopes are not used during the work of the stand, which significantly reduces the cost of the performed research, and does not require the introduction of special measures for the organization of the radiation safety system for personnel.

With the use of the SKIF stand, the dependence of the domestic nuclear power industry on the high-cost services of foreign contractors will decrease.

During the execution of the works, articles were published, and a monograph was written and published: "Certification of coal sorbents of iodine filters of NPP ventilation systems" [12].

ACKNOWLEDGEMENTS

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СТЕНД КОНТРОЛЮ ЙОДНИХ ФІЛЬТРІВ (СКИФ) ДЛЯ СЕРТИФІКАЦІЇ ВУГЛЕЦЕВИХ СОРБЕНТІВ СИСТЕМ ВЕНТИЛЯЦІЇ АЕС

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Надана інформація про створення першого в Україні стенда контролю йодних фільтрів (СКИФ), призначеного для визначення коефіцієнта очищення газоподібних радіоактивних викидів від йоду і метилйодиду сорбентами, що використовуються у системах вентиляції на АЕС України. Обґрунтовано можливість вживання стабільного ізотопу йоду для визначення адсорбційної здатності вуглецевих сорбентів. Проведено розробку, монтаж, запуск і наладку модуля прокачування і модуля вимірювання при використанні повітряних сумішей з метилйодидом на основі стабільного ізотопу йоду ^{127}I. Розроблене програмне забезпечення дозволяє використовувати стенд в автоматичному режимі. Створена інструкція по експлуатації стенда СКИФ і проведена атестація методики визначення коефіцієнта очистки вентильованого повітря від йодистого метилу вугільними сорбентами в рамках стандарту підприємства.