SECTION 2
PHYSICS AND THE TECHNOLOGY OF CONSTRUCTION MATERIALS

UDK 621.533

DETERMINATION OF THE COEFFICIENT OF IODINE ABSORPTION BY MATERIALS OF CARBON ABSORBERS OF VENTILATION SYSTEMS OF NPP USING STABLE ISOTOPES OF IODINE

National Scientific Centre “Kharkiv Institute of Physic and Technology”, Kharkov, Ukraine
E-mail: levenets@kipi.kharkov.ua

Nuclear-physical method of determining of the coefficient of absorption of iodine by carbon materials using stable isotopes of iodine is presented. Equipments are designed and created for the pumping of air with iodine vapors and for the measuring of iodine content. The processes of dynamic sorption of iodine in industrial carbon adsorbents and the possibility of determining the iodine content by nuclear-physical methods are studied, and the metrological characteristics of X-ray radiometric method are presented. Application of method allows carrying out the certification of carbon adsorbents for gas-cleaning systems and improving the safety of nuclear power plant (NPP) operation.

INTRODUCTION

The improving of safety of exploitation of NPP is one of basic directions of development of nuclear energetic of Ukraine. The system of gases purification, which prevents the outbreak of radioactive pollution in air of atmosphere is the important factor determining of radioactive and ecologic safety of the working of reactor. The great requirements for such systems – a high effective purification of air (99.0...99.95%) and dependability of the working. The development of new carbon filters and necessity of control and determination of term of their availability for the using in system of ventilation of NPP need of methods of determination of coefficient of absorption of iodine by carbon sorbents. The available method of determination of coefficient of absorption of iodine by carbon sorbents is based on the using of radioactive iodine (131I) and their compounds (methyl-iodine)) [1]. The using of such methods requires the additional steps of radioactive safety and license for the working with open radioactive sources, so it is necessary to provide the biological protection from effect of methyl-iodide which have strong carcinogenic properties. Now nuclear energetics of Ukraine haven't of such techniques.

The aim of work is the creation of nuclear-physics methods of determination of coefficient absorption of iodine by carbonic materials of air filters of system ventilation of NPP using stable isotopes of iodine.

EQUIPMENT AND EXPERIMENT

We believe that the adsorption features of the carbon matrix do not depend on the isotopic composition of iodine in the gas mixture. Therefore, to simplify the hardware for realization of analysis of iodine content the method of determining of the coefficient of the absorption of iodine by carbon materials is based on using of pairs of stable isotopes of iodine. Experimental equipment presupposes the existence of two modules:

1 – pumping the air with iodine vapors through a analogue of carbon absorber;
2 – measuring the iodine content in the absorbent material of carbon filter.

The module of the pumping. To study the adsorption of iodine vapors in a dynamic mode of pumping module has been created (Fig. 1). This module simulates the operation of an absorber AU-1500, which is used in nuclear power plant ventilation system.

The specific flow of ventilation air module was 0.48 m³/(s·m²) the thick of layer of absorber was 300 mm. Layout of absorber consisted of 6 sealed sections of 48 mm in diameter and 50 mm in length and was used to obtain data about distribution of adsorbed iodine in activated carbon. Density of iodine in pumped air can be varied by temperature of generator of iodine vapors and by value of air flow through it in the range of \(4.7 \times 10^{-3}...7 \times 10^{-6}\) vol.%, which corresponds to \((0.55...4.6) \times 10^{-4}\) g/m³. Time of pumping of air through the layout of absorber is fixed.

The adsorption capacity of the adsorbent was estimated based on exploitation of AU-1500 in normal operation for 30 years. Thus, when the air stream through an absorber is 1500 m³/h over 30 years of continuous operation through AU-1500 passes \(3.9 \times 10^8\) m³ of air in which the volume content of iodine is \(2 \times 10^{-16}\) vol.%, or \(7.8 \times 10^{-10}\) m³ iodine vapor, which corresponds to \(\sim 8 \times 10^{-6}\) g of iodine. Density of iodine in air flow is about 0.005 g/m³ at nominal air flow through the layout of generator of iodine vapor at room temperature. During the hour about 3 m³ of mixture of air with iodine vapors is pumped thru the layout of absorber, i.e. \(\sim 0.015\) g of iodine. Thus, the layout has a huge dynamic range (about 4 orders of magnitude) relatively to weight of iodine in the pumped gas mixture and allows for a reasonable amount of time of pumping (some minutes) to simulate the operation of the carbon adsorbent during 30 years of its work in the ventilation system of NPP in normal mode.
Module of content iodine measurement. The module of determination of iodine content (MDIC) was created during this work (Fig. 2). At determining radioactive iodine isotopes there are often used radiometry, neutron activation, gamma-activation and mass spectrometry analyzes [6]. However, in this embodiment, when you need a highly sensitive determination of iodine content in the carbon matrix it is preferable to use the methods of elemental analysis based on excitation and detection of the characteristic X-ray emission of iodine [7, 8].

To choose the most optimal variant, experimental studies have been carried out of analytical and technical features of a number of techniques and devices using for analysis of the content of iodine in carbon. This techniques are based on such methods: X-ray radiometric (XRA), X-ray excitation by accelerated ions (PIXE), X-ray fluorescence (XRF). For these proposes we used: apparatus for elemental analysis Elean [9]; Analytical Nuclear Physics Complex SOKOL [10]; layout of settings for XRA MDIC, which were designed and created in KIPT. Analysis of the obtained data showed, that the most preferred for the solving of assigned task is the using of XRA. The influence of the characteristics of primary (exciting) radiation on the X-ray spectrum was studied. and the type of radioactive source $^{241}$Am was selected during the experimental work. According to the results of researches the response functions of 10 detection units with different types of crystals type of detectors of X-ray emission in the range of 30...40 keV was selected. X-ray optical scheme of installation of MDIC and the characteristics of absorbing filters and collimators were identified. Operating parameters of the spectrometric channel were determined.

Quantitative determination of iodine was performed by external standard method using a calibration curve. Calibration samples at the content of iodine in carbon: 0.5; 0.1; 0.05; 0.01; 0.005; 0.001 wt.% were used to fit the calibration curve. Fig. 3 shows the X-ray spectra from calibration samples excited by radioactive source "IGIA-1M-5" with the isotope $^{241}$Am. Registration of radiation was carried by Ge(HP)-detector with a resolution of 250 eV at 6.4 keV line. The fitting of the obtained values by a linear function according to method of least squares was carried. The correlation coefficient between the area of analytical peak and concentration was $r_{xy} = 0.99863$.

**Fig. 3. X-ray spectra from calibration sample excited by radioactive source $^{241}$Am. The detector Ge(HP)**
number of granules (the average length of the granules ~ 3.7 mm) is about 720 pieces, and their total surface, which is bordered by a stream of air ~ 250 cm². Thus, the carbon absorber in the layout can be seen (Fig. 4) as an isotropic porous medium, which almost has no resistance when air is pumped through.

![Fig. 4. Layout of absorber for dynamic pumping of iodine vapors](image)

When the length of the layout is equal L, the cross-section layout absorber - S, the linear flow rate of air, the concentration of iodine at the beginning of the layout - C₀, iodine concentration in the volume of coal at a distance x from the absorber C(x), the rate of sorption of iodine by carbon from the corresponding portions of air can be determined by the formula:

\[ \frac{dC(x)}{dt} = -AC(x), \]

where A – relative coefficient of iodine sorption by coals from the relevant portions of air. When the linear flow rate of air is \( V = \frac{dx}{dt} \) and taking into account the fact that \( V = \frac{B}{S} \) (B is volume pumping rate), we obtain

\[ \frac{dC(x)}{C(x)} = -\frac{AS}{B} dx. \]

After integration of the equation we obtain the dependence of the concentration of iodine from the absorber flow rate of air along the length of the layout:

\[ C(x) = C₀ \cdot \exp \left( -\frac{ASx}{B} \right). \]

If we introduce \( k = \frac{A}{B} \) the coefficient of volume absorption, and the number of absorption sections is n, so the distribution of iodine can be expressed by the formula:

\[ C(n) = C₀ \cdot \exp \left( -\frac{kn}{B} \right). \]

When n=6, taking into account that the absorption coefficient is equal \( K_{abs} = \frac{C(0)}{C(6)}, \) we obtain the following equation:

\[ \ln \frac{1}{K_{par}} = \ln \frac{C(6)}{C(0)} = -6k. \]

According to the recommendations of IAEA [11] and the standard US and EU [12] the minimal value of the ratio of iodine content in the air at the inlet of the absorber layout to its content at the outlet should be not less than 100.

So, when the ratio is equal to 4.605, it can be argued that the absorber is suitable for use in air treatment systems of nuclear power plants.

**MEASUREMENTS**

The measurements were carried out for a number of carbon adsorbents from different manufacturers: DGF2, Carbo tech (Germany), Norit RKJ (Belgium), SKT-31 (Russia), Electrode-D (Ukraine). These adsorbents were studied because of they have a complex of technological characteristics: high strength to abrasion; acceptable aerodynamic resistance of layer and a high adsorption capacity of iodine and methyl iodide. Preliminary studies were carried out of physical and mechanical properties of sorbents [13], partially, their hardness, coefficient of friction, wind capacity, and so on.

The sorbents were loaded into the module for dynamic pumping (6 sections were loaded). The desired density of iodine vapor in the air stream in the experiments was 0.077g /m³. Time of pumping of the air stream with vapor or molecular iodine through a layout of absorber (τ) was 2, 4, and 6 hours. After the finishing of the pumping granules of sorbent were extracted from the layout of sorption module. From each section the sample weighing about 5 g was selected in which the content of iodine was studied. Determination of iodine content was made in module of measurement.

**RESULTS**

We study the dynamical regimes of pumping iodine vapor using developed module of pumping and so study the metrological characteristics of the module of measurements of iodine content using adsorbents in the market. In the process of studying of features of carbon sorbents the content of iodine in individual granules of sorbents impregnated with iodine was determined. Fig.5 shows the iodine content in the individual granules of sorbents used in ventilation system of NPP. Such variation of the iodine content in the individual granules of sorbent required of ascertainment of the analysis representativeness. The obtained data been shown, that sample with weight about 5 g for these types of carbon absorbers (size factor) is representative for the determination of iodine content. The error of measurement of the concentration of iodine is 0.003...0.007%. Experimental verification of method of determination of purification factor was performed on the carbon adsorbent Norit. The experiments related to the dynamic pumping in the module of vapor of iodine have shown that there is an optimal time of pumping.

Fig. 6 shows a plot of the logarithm of the concentration of iodine (C) as function of the section number (N) constructed by measuring the content of iodine in the substance of each section of layout. For the experimental points a fitting was made by function of the type \( \ln (C) = a \cdot N + b \) (see Fig 6), which was subsequently used to determine the iodine absorption coefficient (Kₚₐ₉). For carbon adsorbent Norit \( K_{par} = 99.96 \), what is consistent with regulations.
CONCLUSIONS

The nuclear-physical method was developed of the determination of the coefficient of iodine absorption by carbon adsorbents designed for gas cleaning system of NPP. Proposed method based on the using of stable isotopes of iodine.

The layout of the stand for the implementation of the method was developed, built and tested. The stand includes the module a dynamic pumping of iodine vapor and the module of determining of iodine in adsorbent using XRA based on the source $^{241}$Am. A number of researches were made aimed at the optimizing of the modes of dynamic pumping of mixture air with iodine vapors and metrological characteristics determination of iodine adsorption.

REFERENCES


Article received 25.11.2015
ОПРЕДЕЛЕНИЕ КОЭФФИЦИЕНТА ПОГЛОЩЕНИЯ ЙОДА УГЛЕРОДНЫМИ МАТЕРИАЛАМИ АДСОРБЕРОВ СИСТЕМ ВЕНТИЛЯЦИИ АЭС С ИСПОЛЬЗОВАНИЕМ СТАБИЛЬНЫХ ИЗОТОПОВ

В.В. Левенец, В.И. Соколенко, Э.И. Винокуров, А.Ю. Лонин, А.П. Омельник, Р.М. Сибилева, А.А. Щур

Представлена ядерно-физическая методика определения коэффициента поглощения йода углеродными материалами с использованием стабильных изотопов йода. Разработаны и созданы модули прокачки и измерений содержания йода. Исследованы процессы динамической сорбции йода на промышленных углеродных адсорбентах, изучены возможности определения содержания йода ядерно-физическими методами и представлены метрологические характеристики рентгенорадиометрического метода. Применение методики позволит провести сертификацию углеродных адсорбентов систем газоочистки и улучшить безопасность эксплуатации АЭС.

ВИЗНАЧЕННЯ КОЄФІЦІЄНТА ПОГЛІНУННЯ ЙОДУ ВУГЛЕЦЄВИМИ МАТЕРІАЛАМИ АДСОРБЕРІВ СИСТЕМ ВЕНТИЛЯЦІЇ АЕС З ВИКОРИСТАННЯМ СТАБІЛЬНИХ ІЗОТОПІВ

В.В. Левенець, В.І. Соколенко, Е.І. Вінокуров, О.Ю. Лонін, О.П. Омельник, Р.М. Сібілева, А.О. Щур

Представлена ядерно-фізична методика визначення коефіцієнта поглинання йоду вуглецевими матеріалами з використанням стабільних ізотопів йоду. Розроблені і створені модулі прокачування і вимірювання вмісту йоду. Досліджено процеси динамічної сорбції йоду на промислових вуглецевих адсорбентах, вивчено можливості визначення вмісту йоду ядерно-фізичними методами і представлена метрологічні характеристики рентгенорадіометричного методу. Застосування методики дозволить провести сертифікацію вуглецевих адсорбентів систем газоочистки і поліпшити безпеку експлуатації АЕС.