SECTION X. CHEMISTRY

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Radioiodine Sorption Using UVIS© Silver-Containing Carbon Material

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Abstract

This paper presents a study of sorption capacity of carbon fiber materials impregnated with silver nitrate relative to radioactive methyl iodide. The capacity of the test material frontal layer is defined, and purification degree of steam-air flow from radioiodine is calculated.

Keywords: radioiodine sorption, iodine-131, radioactive methyl iodide, silver-containing sorbents.

Effective air purification in working premises of nuclear industry is an urgent task because of increasingly stringent requirements to gas-aerosol emissions. Removal of radioactive forms of iodine is usually carried out in two stages. First, aerosol component of emissions is captured, and then gaseous compounds of iodine are captured using absorption with impregnated sorbents having large specific surface area. In some cases, radioiodine removal from air flow is carried out by means of combined filters containing layers of filtering materials for coarse and fine cleaning from aerosols and also absorbing layers made of coal or other adsorbents. Carbon fabric based on hydrate cellulose fibers – UVIS© – can be one of these materials.

In this paper, investigation of radioactive methyl iodide sorption using UVIS© material impregnated with various amounts of silver nitrate is described. The aim of the study was to assess suitability of this kind of sorbents for use in industrial iodine purification devices of FAI-3000 type and its analogues.

Impregnation of carbon fabric was carried out with aqueous alcohol solutions of silver nitrate, followed by drying. The final concentration of AgNO_3 in sorbents was from 1 to 8wt.%. It can be noticed that silver nitrate is distributed unevenly over the fibers with a diameter of 5-7 microns (Fig. 1). There are some areas where the impregnant concentration is increased. AgNO_3 agglomerates range in size from 0.4-0.6 μm to 2-3 μm .

Figure 1. Samples of the sorbent containing 4wt.% AgNO_3 : a - increase 2000; b - increase 20000

Sorption capacity of the obtained samples was evaluated using the control iodine stand of D. Mendeleev University of Chemical Technology of Russia under the following conditions: gas flow temperature – $30.0 \pm 0.1^\circ\text{C}$; relative humidity – $90.0 \pm 1.5\%$. Samples of the carbonic fiber with a diameter of 50 ± 1 mm and thickness of 0.5 mm were placed in turn into the column of the stand; at the same time, methyl iodide vapors labeled with iodine-131 isotope were supplied together with the steam and air mixture. Radioiodine concentration in the air flow was 60-80 mg/m³. The gas flow velocity in the column section was 2.3 cm/s that corresponds to linear velocity in the industrial gas cleaning apparatus. After the test, the column with the sorbent was dismantled and CH³131I radioactivity distribution by the sorbent layers was evaluated for 364 keV energy line using gamma-x-ray spectrometer. Degree of radioiodine removal from gas-stream mixture was calculated on the basis of the radioactivity distribution:

$$E = \frac{A_1}{\Sigma A} 100\% \quad (1)$$

where A_1 is radioactivity of CH³131I caught with carbon fiber material with thickness of 1 mm (cps); ΣA is total radioactivity of radioiodine supplied into the system (cps).

Capacity of the material frontal layer was defined based on data on its radioactivity and radioactivity of the reference source in which specified amount of labeled methyl iodide was placed. Geometry of the samples was the same.

Fig. 2 shows kinetics of the radioactive methyl iodide accumulation in frontal layer of carbon fiber material.

The experimental point vector was described by $f = A \cdot (1 - e^{-km})$ functions obtained by the least squares method, where A is working capacity of the sorbent; m is amount of the radioiodine supplied. As a result, it was found that dynamic radioiodine capacity is not additive. For samples containing 1%, 2%, 4%, 8% nitrate of silver, limiting amount defined by this method was 0.25, 0.30, 0.31, 0.49 mg/m² of methyl iodide. The curves shape makes it possible to suggest that, at the initial moment, chemical reaction rate is higher in samples impregnated with less amount of silver nitrate (1% and 2%). Apparently, this is due to the distribution of crystals over the material surface, as well as due to the crystal size.

Results of purification degree calculation for the case of gas flow purification from radioiodine using carbon fiber material with a thickness of 1 mm versus amount of CH³131I supplied to the system is given in Table. 1. There is an obvious tendency to reduction of the purification degree in the course of the impregnant chemical reaction in the sorbent.

Standard requirements for sorption-filtering materials used in iodine purification devices are as follows: dynamic iodine capacity – not less than 0.7 mg/m²; gas flow purification degree – not less than 90%. According to Table 1, all samples of silver-containing carbon fiber material meet the second requirement up to amount of methyl iodide supplied into the system of ~6 mg.

Figure 2. The dependence of the capacity of the frontal layer of impregnated carbon fiber on the amount of the $\text{CH}_3^{131}\text{I}$

The first requirement can be fulfilled if number of sorption filter material layers is increased. It is obvious that, in this case, total degree of purification will not be reduced. However, with this approach, it is necessary to control overall hydraulic resistance of the resulting sorption assembly, since the costs of ventilated air pumping play an important role in the operation of the entire gas cleaning system of the nuclear power plant.

Table 1.

The calculated values of the degree of purification (E)

Silver nitrate content with sample							
1%		2%		4%		8%	
Feed amount $\text{CH}_3^{131}\text{I}$, mg	E , %	Feed amount $\text{CH}_3^{131}\text{I}$, mg	E , %	Feed amount $\text{CH}_3^{131}\text{I}$, mg	E , %	Feed amount $\text{CH}_3^{131}\text{I}$, mg	E , %
3.1	95.44	3.2	95.70	1.5	97.02	2.8	95.29
5.9	90.04	5.7	93.86	5.2	91.24	5.4	91.27
9.3	76.91	9.0	82.22	8.7	83.69	8.6	87.94
13.0	63.10	13.5	68.72	12.6	73.32	12.5	80.89
-	-	18.4	52.45	-	-	16.9	73.25

According to the study results, it was found out that, with the increase in the AgNO_3 concentration, limiting radioactive methyl iodide capacity of sorption-filtering materials is not additive, and that all tested materials remove radioactive methyl iodide from the gas flow with efficiency above 90% up to 6 mg of total $\text{CH}_3^{131}\text{I}$ amount supplied to the system.

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