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Study on adsorption performance of coal based activated carbon to radioactive iodine and stable iodine

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ABSTRACT

Nuclear power plant, nuclear reactors and nuclear powered ship exhaust contains a large amount of gaseous radioactive iodine, and can damage to the workplace and the surrounding environment. The quantitative test to remove methyl iodide and the qualitative test for removing stable iodine were investigated using the impregnated coal-based activated carbons and coal-based activated carbons as adsorbents. The research conducted in this work shows that iodine residues were under 0.5 μg/ml after adsorption treatment and the decontamination factor of the coal-based activated carbon for removing the stable iodine was more than 1000, which can achieve the purpose of removing harmful iodine, and satisfy the requirement of gaseous waste treatment of nuclear powered vessel and other nuclear plants.

1. Introduction

Equipped with nuclear propulsion plant, and taken it as the main power plant of vessel is nuclear powered vessel. Similar to land-based nuclear power plant system, nuclear reactors of vessel nuclear power system will produce radioactive gaseous waste. Of the radioactive wastes, radioiodine such as elemental iodine, organic iodide and hypoiodous acid are the most important nuclides due to the volatilization and the significant radiological effects on human body and environment (Hou et al., 2007; Woo, 2013). So emphasis has been placed on the reduction of radioactive gaseous waste released from nuclear facilities for the several years (Choi et al., 2001). Removing radioactive iodine from nuclear power plant gaseous waste, especially its organic forms, is an important problem in the operation of nuclear power plants (Ampelogova et al., 2002). Monitoring and control of stable iodine and radioactive iodine pollution has been the focus of attention.

Radioactive iodine mainly exists in gas and liquid. Gaseous radioactive iodine trapping methods are varied, mainly be divided into four categories: precipitation, dry dedusting, liquid absorption and solid adsorption method. Ali et al. (2013) studied the iodine removal efficiency in a self-priming venturi scrubber, and got the best operating condition. Liquid radioactive iodine capturing methods mainly include metal adsorption and adsorbent adsorption (Kulyukhin et al., 2011). In this work, we study gaseous iodine adsorption. Generally, solid adsorption is studied more (Liu and Liu, 1996), such as inorganic capture material (Ramos et al., 2013; Funabashi et al., 1995; Tang et al., 1987; Ye et al., 1991), activated carbon (Yue et al., 2013) and carbon-fiber (Ampelogova et al., 2002). Activated carbon is a porous carbon material, and possesses a high adsorption capacity. As adsorbent, it's widely used in the purification of gases and liquids (Angın et al., 2013). In nuclear plants, activated carbons are widely used to capture radioactive iodine compounds at different systems. For the adsorption of methyl iodide, the activated carbons are usually prepared from coal or coconut shells (Park et al., 2001). At present, for the nuclear industry, the impregnated activated carbon based coconut shell is the most effective (Gonzalez-Garcia et al., 2011), and can meet the demand of nuclear grade of carbon removing iodine. However, the price of coconut shell activated carbon is expensive, and its airflow resistance is relatively larger (Yue and Luo, 2012). Li et al. (1987) used different activated carbon and impregnant to adsorb gaseous radioactive iodine, and studied the influence of different experimental conditions on the adsorption efficiency. Huang et al. (1988) selected several types of coal-base activated carbon and impregnant to examine for their main physical and chemical performance. Most of their physical properties can satisfy the requirements of the nuclear grade charcoals. Also, there are some

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problems regarding the use of activated carbon in nuclear power plant, as: point of ignition is low; providing explosion risk in case it absorbs a large amount of nitrates; releasing radioiodine after long continuous times. The price of coal-based activated carbons produced in Shanxi is low, which is suitable for industrial mass production applications, and it has some excellent quality, such as low sulfur content, high hardness (>97%) and higher ignition (420 °C), so it can be used as the adsorbent replaced the expensive shell carbon to remove iodine.

The quantitative test to remove the methyl iodide utilizing the iodine adsorber and the impregnated coal-based activated carbon produced by Shanxi Xinhua Chemical Factory was did, then the qualitative test of the coal-based actived carbon for removing stable iodine on the base of the first test was carried out. The research conducted in this work shows that the decontamination factors (defined as the ratio of the initial activity to the final activity) of the coal-based actived carbon for removing stable iodine reached more than 1000, so it can satisfy the need of project application, and is very important to deal with the gaseous waste of nuclear powered vessel and other nuclear facilities.

2. Materials

2.1. Absorbents

The actived carbons adopt coal-based activated carbon DX-30 impregnated in KI (black cylindrical particles, high-quality anthracite, particle size is 1.0–4.0 mesh), which are produced by Shanxi Xinhua Chemical Factory. Columnar coal-based activated carbon has a reasonable pore structure, good adsorption properties, high mechanical strength, easy to repeated regeneration, and low cost characteristics. Its physical and chemical analysis properties are shown in Table 1.

2.2. Instruments

The iodine adsorber is LX-100, produced by Shanxi Xinhua Chemical Factory. LX-100 iodine adsorber belongs to the folding iodine adsorber, and is composed of the metal shell, activated carbon adsorbent layer, sealing materials and other accessories.

3. Theory and methods

The principle of removing iodine by using KI impregnated activated carbon is mainly isotope exchange (Jia, 1992):

$$CH_3^{131}I(g) + K^{127}I \iff CH_3^{127}I(g) + K^{131}I \tag{1}$$

The isotope exchange is a reversible process, so there should be obvious excess iodine in the adsorption phase in order to guarantee comparatively high efficiency of the removal of radioactive iodine and methyl iodide. The process of removing organic iodine consists of significantly larger number of steps than the physical adsorption process alone (Kovach, 2006), for example, active form diffuse to the grain surface, active form diffuse into the pores, physical adsorption of active form, isotopic exchange, desorption of inactive form from the surface, diffusion of stable form from pores, and diffusion of stable form into the gas phase.

Therefore, if the decontamination factors of the impregnated activated carbon for removing the radioactive iodine and methyl iodide can be more than 1000, the decontamination factor of the non-impregnated activated carbon for removing stable iodine will only be higher than it. Because the removal of the radioactive iodine still need isotope exchange except absorption step, however removing stable iodine only need an absorption process. So this experiment mainly adopted the method of nuclear power plant removing iodine, and carried on the adsorption characteristic experiment of the DX-30 coal-based actived carbon. As long as we can prove that the coal-based actived carbon can meet the purification demand for the radioactive iodine and also prove it can meet purification demand for the stable iodine.

Iodine removal efficiency adopts decontamination factors-DF for the methyl iodide (Sinha et al., 1997), defined as the ratio of the initial activity to the final activity.

$$\mathrm{DF} = \frac{A_0}{A_t} \tag{2}$$

where A_0 is the initial activity of ^{131}I, A_t is the activity at time t after iodine adsorption, and the unit is MBq.

4. Experimental

4.1. Experiments of impregnated coal-based actived carbon for removing methyl iodide

We carried on the characteristic experiment of the coal-based activated carbon, which made use of the complete iodine adsorption adsorber LX-100 produced by Shanxi Xinhua Chemical Factory, and the loaded absorbent is the impregnated KI coal-based activated carbon, which is produced by Shanxi Xinhua Chemical Factory. According to France standard AFNOR NFM62-206, the DF for methyl iodide in normal and limit operating temperature were tested. The flow chart of the experiment is shown as Fig. 1.

In the experiment, the methyl iodide gas generator produced methyl iodide marked ^{131}I, and poured into the piping continuously for 30 min. The gaseous iodine sampling instruments were set up in upstream and downstream of the iodine adsorber, and there were first-class filter and two-class activated carbon box. The filter was set up in the entry of the sampling instruments to capture the aerosol of methyl iodide. The first class activated carbon box was used for capturing methyl iodide from the sample gas, while the second class was the supernumerary carbon bed. The gas flow was 3 m^3/h while sampling, and the upstream and downstream sampling time were 45 min and 60 min respectively. After sampling, we tested the activity of methyl iodide adsorbed in the upstream and downstream carbon box by the USA BNC SAM-940 type NaI portable γ spectrometer, and then calculated out the DF of iodine adsorption vessel removing methyl iodide.

Different air stream conditions and sampling conditions were established to determine the performance of the coal-based actived carbon, and the experiment of the complete adsorption

Table 1
Physical and chemical properties of DX-30 actived carbon used in the experiment.

Analysis items	Test data	Analysis items	Test data
Iodine value (mg/g)	>1000	Total pore volume (cm^3/g)	>0.8
Intensity (%)	>92%	Adsorption of residual chlorine (%)	⩾85%
Specific surface area (m^2/g)	>1000	Filling density (g/$cm^{3)}$	0.45–0.55
Methylene blue (mg/g)	120–150	Particle size (mesh)	1.0, 1.5, 2.0, 3.0, 4.0