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Research Paper

Efficiency of Moso Bamboo Charcoal and Activated Carbon for Adsorbing Radioactive Iodine

Preventing radioactive pollution is a troublesome problem but an urgent concern worldwide because radioactive substances cause serious health-related hazards to human being. The adsorption method has been used for many years to concentrate and remove radioactive pollutants; selecting an adequate adsorbent is the key to the success of an adsorption-based pollution abatement system. In Taiwan, all nuclear power plants use activated carbon as the adsorbent to treat radiation-contaminated air emission. The activated carbon is entirely imported; its price and manufacturing technology are entirely controlled by international companies. Taiwan is rich in bamboo, which is one of the raw materials for high-quality activated carbon. Thus, a less costly activated carbon with the same or even better adsorptive capability as the imported adsorbent can be made from bamboo. The objective of this research is to confirm the adsorptive characteristics and efficiency of the activated carbon made of Taiwan native bamboo for removing ^{131}I gas from air in the laboratory. The study was conducted using new activated carbon module assembled for treating ^{131}I-contaminated air. The laboratory results reveal that the ^{131}I removal efficiency for a single-pass module is as high as 70%, and the overall efficiency is 100% for four single-pass modules operated in series. The bamboo charcoal and bamboo activated carbon have suitable functional groups for adsorbing ^{131}I and they have greater adsorption capacities than commercial activated carbons. Main mechanism is for trapping of radioiodine on impregnated charcoal, as a result of surface oxidation. When volatile radioiodine is trapped by potassium iodide-impregnated bamboo charcoal, the iodo-compound is first adsorbed on the charcoal surface, and then migrates to iodide ion sites where isotope exchange occurs.

Keywords: Adsorption efficiency; Air pollution; Bamboo activated carbon; Moso bamboo charcoal; Radioactive iodine

Received: January 11, 2010; *revised:* April 28, 2010; *accepted:* June 3, 2010

DOI: 10.1002/clen.201000012

1 Introduction

Preventing radiation pollution is an urgent but troublesome problem worldwide. Radioactive materials in our environment may enter human body to cause serious and irreversible health-related problems. Since their discovery about a century ago, radioactive materials have been extensively used in medicine, agriculture, industry, and even our daily living. Overall, the radiation has a negative image in the mind of most people; it is always blamed for all types of cancers. In medicine, radiation has been a convenient treatment tool for more than 80 years. With advances in science and technology, the radiation treatment is becoming a specialized science for treating the various cancers such as head and neck cancer, cervical cancer, breast cancer, esophageal cancer, colorectal, and prostate cancer [1].

Thyroid cancer, which often occurs in female patients, is usually removed surgically. The remaining thyroid is irradiated using ^{131}I with an activity (30–200 mCi) for a total thyroid ablation. Generally, the safety of ^{131}I use in medicine and proper disposal of the waste ^{131}I is of remain important concerns.

Because of its superb adsorptive characteristics, activated carbon has been widely applied in various industries for purifying and de-coloring materials, and removing toxicity from various products. It has also been extensively used for treating wastewater and air emission; the consumption of activated carbon is greater for nations that enforce more stringent environmental protection laws and regulations. In recent years, the total annual consumption of activated carbon is around 400 000 tons with 41% consumed in the US, 22% consumed in Western Europe, and 23% consumed in Japan; the total global production of activated carbon is 520 000 tons. In Taiwan, various air and water pollution control and prevention laws have gradually passed and enforced, the consumption of activated carbon is expected to rise annually. According to the 2000 Republic of China custom statistics, the annual import of activated carbon in that year exceeded 10 000 tons.

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Abbreviations: KI, potassium iodide.

The activated carbon is a solid that has high specific surface area with high porosity; its pores provide strong capacity to adsorb and separate solute from solvent [2]. The porous structure is defined by pore size distribution. Based on the definition of International Union of Pure and Applied Chemistry (IUPA), porous structures can be classified into three categories. The first category includes those that have small pores to allow one or two molecules to be adsorbed per pore with strong interaction between the adsorbed molecules and the pore wall. The secondary category has mesopores that may demonstrate capillary phenomena and characteristics of hysteresis loops under relatively high pressure [3, 4]. The third category consists of macroporal sizes with relatively lower ratio of surface area to porosity. Although the particles with macropores have low specific surface area, the pore can serve as channels between meso- and micropores. In the filters of nuclear establishments, carbon materials impregnated with either potassium iodide (KI) or triethylenediamine (TEDA) or both is used for the trapping of radioiodine, one of the major fission products released during a nuclear accident [5]. The carbon materials come from two sources, bamboo charcoal and bamboo activated carbon by self-made and coconut and anthracite activated carbon of commercial materials.

The objective of this research is to evaluate the feasibility of using bamboo activated carbon for removing radioactive pollutants, and to compare the effectiveness of the bamboo activated carbon with commercial available coconut and anthracite activated carbon. Beside, try to effect of adsorbent with impregnated 5% KI on various carbon materials. A novel design of air pollution treatment module that is fast, convenient and can be applied on-site is proposed in this research for carrying out the evaluation. This module allows effective changes of the testing adsorption material; it can also be operated in series or parallel and can be immediately applied on-site after laboratory evaluation.

2 Materials and methods

2.1 Air sampling

The activated carbon particles to be evaluated were packed in the air pollution treatment module, which were connected either in series or parallel in a vertical position to assure close packing of the media. A filter paper was placed at the front end to filter out the atmospheric particles. Air sampling was done in the ^{131}I laboratory where most of the ^{131}I particles exist in the air as I_2. The connected modules were then hooked to an air sampler; air was withdrawn through the adsorption media continuously.

After sampling the air, the adsorption media packed in various layers were removed and placed in sealed acrylic containers. The weight for each adsorbent is measured and the samples were prepared for the following radioactivity measurements.

2.2 ^{131}I Activity measurement

The adsorbents from each layer were placed in the gamma spectrometer for determination of the ^{131}I activity. The characteristic gamma rays of 364 keV emitted from ^{131}I were detected using a high-purity germanium detector and gamma spectra were analyzed using software to determine the measured ^{131}I activity. The specific activity in Bq/g was used to minimize the errors caused by the differences of adsorbent placed in each layer. The activity of ^{131}I is in unit of Becquerel (symbolized by Bq).

2.3 Experimental system and materials

2.3.1 Experimental system

The new air treatment module units may be connected in series or parallel for air sampling. The number of total modules connected in series will be further investigated in order to obtain data for implementing the module in a full-scale power plant. The experimental system is schematically shown in Fig. 1.

2.3.2 Materials

The ^{131}I gas, which contains predominantly I_2, and the specifications of all four adsorbents, *i.e.*, bamboo activated carbon, bamboo carbon, anthracite activated carbon, and coconut activated carbon, are listed in Tab. 1. All bamboo activated carbon used in this research has been developed by the authors' research team. There are seven steps involved in making the bamboo activated carbon [6] as shown in Fig. 2. Procedures for achieving bamboo activated carbon particles are briefly discussed below.

2.3.2.1 Carbonization

The 3–4-year-old Moso bamboo was subject to high temperature (550°C) under conditions of limited air for the organic components to be thermally decomposed. After the organic components are oxidized or evaporated, the residual carbon polymerizes to form a huge carbon skeleton.

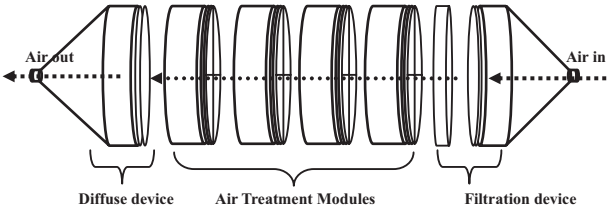

Figure 1. Novel module about air treatment.

Table 1. The characteristics of four adsorbent materials

Material	SSA (m^2/g)[a)]	Mesh[b)]	Diameter (mm)
Anthracite activated carbon	700	10–30	1.6–0.6
Coconut activated carbon	900	8–12	2.0–1.4
Bamboo activated carbon	900	16–32	0.9–0.5
Bamboo charcoal	400	2–8	5.0–2.36

[a)] Specific surface area (BET).
[b)] Particle size.

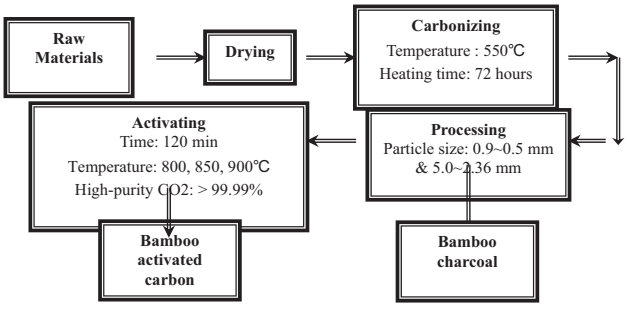

Figure 2. Processes of made bamboo charcoal and activated carbon of bamboo.

2.3.2.2 Activation

The numerous holes and pores created during the carbonization process in the carbon structure are easily plugged by the adsorbed materials. Thus, the carbon must be activated to increase its specific surface area and porosity, and the adsorptive capacity.

In this research, 20 g of bamboo carbon (16–32 meshes) were placed in a super-high temperature vacuum carbon activating furnace to activate the carbon. After activated for 120 min at three different high temperature levels (800, 850, and 900°C, respectively), and import the activation of gases to high-purity (more than 99.99%) of CO_2. The activated carbon was further dried in an oven (103 ± 2°C) until it was absolutely dry as determined by its weight.

2.3.2.3 Sampling with KI-impregnated bamboo charcoal

A commercially available KI-impregnated carbon (nuclear powerlevel) manufactured by impregnating the charcoal of *Moso* bamboo with 5 wt% KI was used. It has particle size range of 2.36 to 5 mm (BS two to eight nominal meshes). The KI-impregnated charcoal was predried overnight in an oven of 110°C and then cooled in a desiccator.

The mechanism for the trapping of radioiodine by KI-impregnated charcoal was investigated by loading ^{131}I onto a bed of charcoal impregnated with KI of the same specific activity.

2.3.2.4 Determined of surface functional groups of carbon

The carbon sample was added to the excess standard base (0.001 N $NaHCO_3$, 0.001 N Na_2CO_3, 0.01 N NaOH, or 0.02 $NaOC_2H_5$) solution, and the acidic surfaces were determined by back-titration with HCl after reaching the equilibrium [7].

2.4 Calculation of the adsorption activity

The efficiency of adsorption is expressed in Bq/g for each layer of adsorbent (higher numbers indicate higher efficiencies. The efficiencies for adsorbing ^{131}I are designated as A1, A2, A3, and A4 for respective layer. Assuming that almost 100% of ^{131}I is adsorbed by these four modules, the adsorption efficiency is defined as the efficiency of the first-layer, can be expressed as:

$$\text{Adsorption efficiency}(\%) = \frac{A_1}{A_1 + A_2 + A_3 + A_4} \times 100$$

3 Results and discussion

3.1 Evaluating the adsorption efficiency for various materials

Under conditions of controlled temperature, adsorption rate, and procedures, the single layer adsorption efficiencies are 61.3% for commercial anthracite activated carbon, 59% for commercial coconut activated carbon, 70.9 for bamboo activated carbon, and 72.3% for bamboo charcoal (Fig. 3).

A summary of the pore size distributions of four carbon materials studied in the paper is shown in Tab. 2. Results show that both bamboo activated carbon and charcoal have more than 70% adsorption efficiency and 10% higher efficiency than commercial activated carbon (Tab. 3). The high adsorption efficiency associated with bamboo activated carbon and bamboo charcoal may be due to the presence of numerous micropores that are effective in adsorbing small non-polar ^{131}I gaseous molecules (Tab. 2). As for anthracite and coconut activated carbons, the pore sizes are between meso- and macrosizes, thus they are as effective for adsorbing relatively large molecules [3, 8].

3.2 Adsorption efficiency for bamboo charcoal and activated carbon

Bamboo charcoal and bamboo activated carbon have similar adsorption efficiencies. However, bamboo charcoal particles contact ^{131}I more easily than powdered bamboo activated carbon ^{131}I [9].

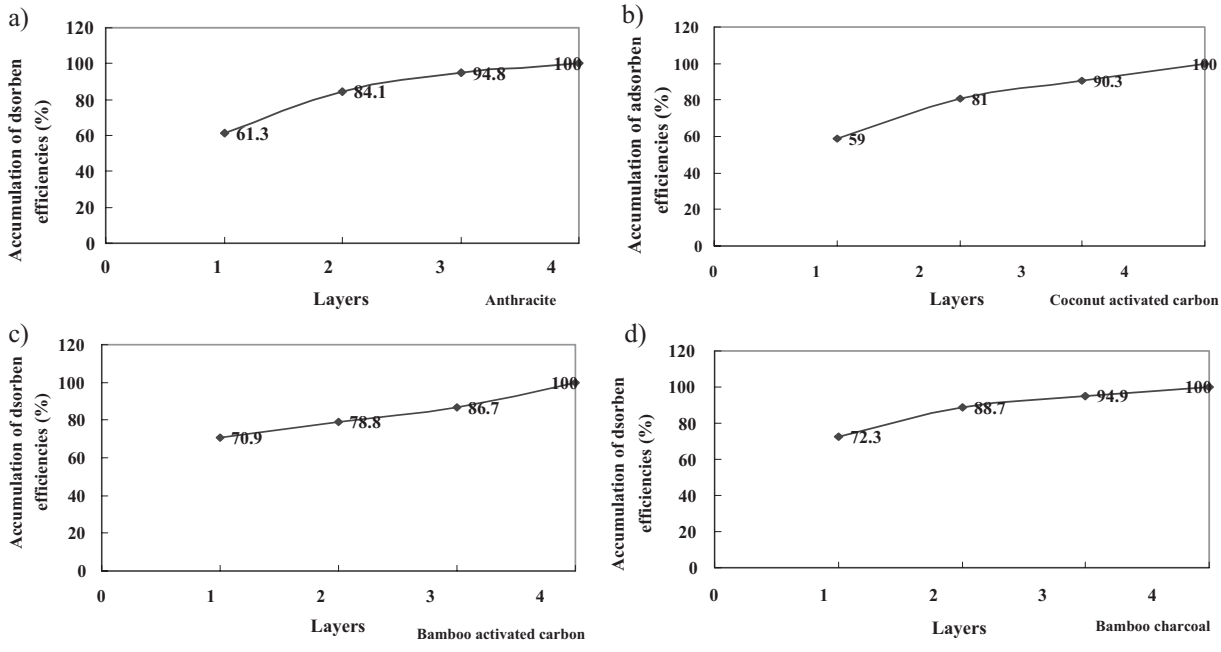

Figure 3. Adsorbent efficiencies of radioactive iodine by various materials.

Table 2. The pore size distribution of variant carbon materials

Pore diameter range (nm)	<6	6–8	8–10	10–12	12–16	16–20	<20	20–80	>80	Total
Anthracite activated carbon Pore volume (%)	29.97	13.31	7.29	7.47	8.17	7.57	73.78	21.73	4.49	100
Coconut activated carbon Pore volume (%)	31.43	11.74	7.05	5.68	6.96	7.65	70.51	22.57	6.92	100
Bamboo activated carbon Pore volume (%)	36.23	13.24	7.07	7.4	7.71	7.24	78.9	17.51	3.59	100
Bamboo charcoal Pore volume (%)	30.63	14.93	8.18	7.9	8.48	7.44	77.56	18.45	3.99	100

Table 3. The distributive doses of ^{131}I at each layer of treatment module

Material	Sample	^{131}I (activation) (Bq/g)	Efficiency of the single layer (%)	Sample weight (g)
Anthracite activated carbon	First layer	1.490 ± 0.041	61.3	22.2
	Second layer	0.555 ± 0.016	22.8	22.2
	Third layer	0.260 ± 0.009	10.7	21.9
	Fourth layer	0.126 ± 0.005	5.2	21.5
Coconut activated carbon	First layer	0.431 ± 0.014	59.0	14.4
	Second layer	0.161 ± 0.006	22.0	14.4
	Third layer	0.068 ± 0.003	9.3	14.6
	Fourth layer	0.071 ± 0.002	9.7	14.1
Bamboo activated carbon	First layer	0.280 ± 0.009	70.9	16.6
	Second layer	0.031 ± 0.002	7.9	17.7
	Third layer	0.031 ± 0.002	7.9	16.3
	Fourth layer	0.053 ± 0.002	13.3	16.5
Bamboo charcoal	First layer	0.033 ± 0.003	72.3	13.6
	Second layer	0.007 ± 0.001	16.4	13.0
	Third layer	0.003 ± 0.000	6.2	13.6
	Fourth layer	0.002 ± 0.000	5.1	13.6

According to literature, under similar temperature, flow velocity and pressure, the adsorptive capacity of activated carbon particles is proportional to the particle size.

The results obtained by Kirubakaran et al. [10] to study fluidized bed and static bed of adsorbent show that when the activated carbon particle diameter increases, the iodine value of the static bed decreases. Thus, pollutants treated with the powdered adsorbent are easier to break through the activated carbon filter bed than those treated in the bed of activated carbon particles. Additionally, for large particle sizes, fluidized bed is more active than static bed; the fluid motion, such as in a fluidized bed, will also enhance the adsorption capacity of large activated carbon particles.

3.3 Effect of adsorbent with impregnated 5% KI

The current use of activated carbon for adsorbing radioactive materials from nuclear power plant effluent was specially processed to be impregnated with 5% KI for enhancing the adsorption capacity. The bamboo charcoal that demonstrates the highest adsorption effectiveness was compared with the 5% KI impregnated activated carbon currently used in nuclear power plants for evaluating their comparative removal efficiencies.

The first layer (2.5 cm of depth) of KI-impregnated Moso bamboo charcoal shows 100% removal efficiency, and raising more than four times of non-impregnated bamboo charcoal, and reduce the 75% of adsorption materials spend. As for first layer of adsorption efficiency higher than 90% about adsorption material of nuclear energy level (RI material), and the adsorption effectiveness on fourth layer had reached 100% (Figs. 4 and 5), but KI-impregnated of charcoal was best interested for high efficiency and low cost objectives by the overall assessment. The results indicate that the main mechanism for trapping of radioiodine on impregnated charcoal is contributed by surface oxidation. When volatile radioiodine is trapped by KI-impregnated activated carbon, the iodo-compound is first adsorbed on the charcoal surface and then migrates to iodide ion sites where isotope exchange occurs [11].

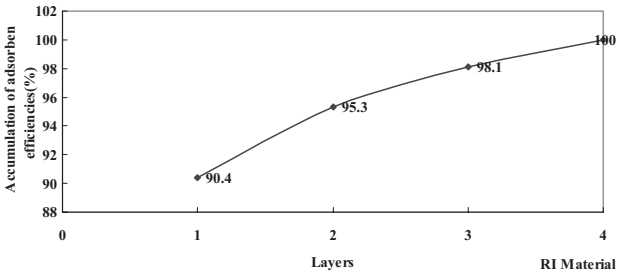

Figure 4. Adsorbent efficiencies of radioactive iodine by RI material.

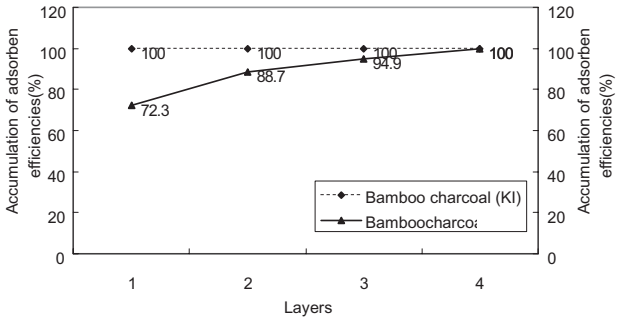

Figure 5. Adsorbent efficiencies of radioactive iodine by bamboo charcoal with immersed KI.

Table 4. The functional group distribution of variant carbon materials

	Carboxyl group (−COOH)	Carbonyl group (−CO−) (mmol g^{-1})	Hydroxyl group (−OH)
Anthracite activated carbon	0.078	0.041	0.103
Coconut activated carbon	0.017	0.016	0.025
Bamboo activated carbon	0.109	0.085	0.115
Bamboo charcoal	0.084	0.083	0.099

3.4 Effect of adsorption efficiency on functional group for surface carbon

The bamboo charcoal and activated carbon have larger amount of basic functional group (carboxyl group, carbonyl group, and hydroxyl group) as compared with coconut and anthracite activated carbons (Tab. 4). In addition, the surface structure of carbon at high temperature condition had some functional groups was decreased and adsorption capacity changed lower [12]. Therefore higher surface functional groups can improve the surface chemical polarity thus increasing the adsorption effectiveness. The basic properties are ascribed to surface basic groups and the π electron system of carbon basal planes, and the basic groups on the surface can increase the electrostatic interactions between the surface and positive charge of KI. The carboxylic group on the surface can release H^+, and it will increase the electrostatic repulsion. So bamboo charcoal and bamboo activated carbon have suitable functional groups for adsorption of ^{131}I and their adsorption capacities are larger than commercial carbons [13].

3.5 Evaluation of the new air treatment module for industrial applications

The testing module proposed in this research consists of three major components: powder filtration component, adsorbent containing component, and gas diffusing layer component. The operational procedures include (1) filling the adsorbent, (2) assembling the power filtering unit, adsorbent-containing component and air diffusion layer component, (3) testing for air tight, (4) testing for adsorption of pollutants for the optimum operating parameters, and (5) calculating the adsorption efficiency. The total time needed to complete these procedures is about 2–3 h. The laboratory adsorption test has confirmed that the new air treatment module proposed in this study will allow direct evaluation of the activated carbon tested. Additionally, this module is easy to assemble and re-assemble, and easy to replace the testing adsorbent for evaluating the adsorption capacity of new material or at a new location. The module is made of aluminum alloy that is highly air tight with the adsorbent filled at different depths or in different layers for studying the optimum efficiency as shown in Tab. 3.

4 Conclusions

The efficiencies of adsorbing ^{131}I from ambient air with four adsorbents, *i.e.*, commercial coconut activated carbon, commercial anthracite activated carbon, bamboo charcoal, and bamboo activated carbon, have been conducted using a new modular design unit. Laboratory results are summarized as follows:

- For a single pass-through adsorption, the order of adsorption efficiencies is bamboo charcoal > bamboo activated carbon > anthracite activated carbon > coconut activated carbon.
- For multiple-layer adsorption, the overall adsorption efficiencies for all four adsorbents tested can reach 100%. The order of adsorption efficiencies for the initial three layers of adsorbent is bamboo charcoal > anthracite activated carbon > coconut activated carbon > bamboo activated carbon.
- Surface oxidation contributes to the trapping of radioiodine on impregnated charcoal. When volatile radioiodine is trapped by KI-impregnated activated carbon, the iodo-compound is first adsorbed on the charcoal surface and then migrates to iodide ion sites where isotope exchange occurs.
- The bamboo charcoal and bamboo activated carbon have suitable functional groups for adsorbing ^{131}I and they have larger adsorption capacities than commercial carbons.
- The air treatment module proposed in this research is easy to operate, relocate, and replace adsorbent material; it will provide industries a convenient tool for conducting on-site evaluation of various absorbents.
- Future research will be directed toward on-site studies for removing radioactive contaminants from nuclear power plant so that more complete and realistic information such as those on material specifications, characteristics, adsorption efficiencies, and cost-effectiveness can be obtained to assist industries in selecting the most efficient and cost-effective treatment scheme for treating radioactive-borne air.

Acknowledgments

This study was funded by Forestry Bureau, Council of Agriculture Executive Yuan in Taiwan. Additional thanks to General Director Yen Jen-Teh of Forestry Bureau, Council of Agriculture Executive Yuan in Taiwan for their assistance throughout this project.

The authors have declared no conflict of interest.

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