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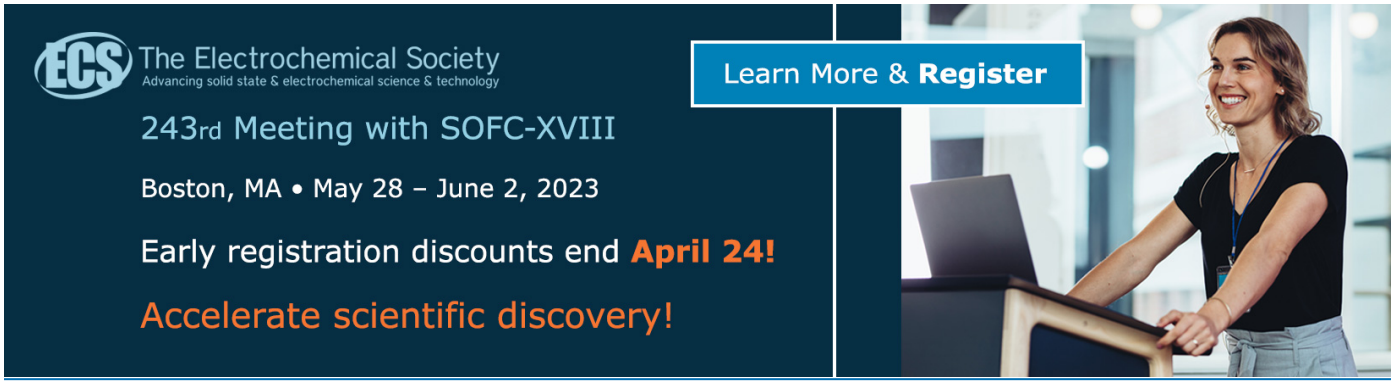

Radioactive Decontamination using Bamboo Activated Carbon for Healthy Environment in Nuclear Medicine

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Abstract. Radioactive wastes by products excreted from radioiodine (RAI) therapy patient waste such as urine, faeces, sweat and puke might risk to radiation contamination if not systematically manage. These wastes can affect human health and environment, thus sustain and systematic management must be strictly considered. In addition, radiopharmaceutical preparations in nuclear medicine risk to radioactive spillage by chances. In this study, new sustainable adsorption technique by using agriculture product was proposed to decontaminate the possibilities of radioactive spillage in RAI therapy where different concentrations of bamboo activated carbon (BAC) was mixed with pure ^{131}I and filtered by using filter paper. Radioactivity for each filtered sample (sediment) was measured using dose calibrator to determine kinetic reactions of adsorbed radioactive substances. The data shows the sediment radioactivity was increased with increased of BAC concentrations. The radioactivity loss after filtration was 76.1% (50 mg/ml), 76.3% (100 mg/ml), 83.5% (150 mg/ml), 80.4% (200 mg/ml), 85% (250 mg/ml) and 68% (control) due to high agglomeration between BAC and ^{131}I. Mixtures with highest BAC concentration was then characterized using FESEM and EDX for morphology and elemental analysis. FESEM image proved there were porous structures on the BAC to attract ^{131}I and other molecules. EDX revealed that ^{131}I and other elements were attracted to BAC layered sheets. This study revealed that BAC performed different capabilities as an adsorbent material under different experimental conditions and has high potential for sustainable radionuclide decontamination agents especially for RAI therapy in ensuring continuous healthy environment for staff, patients and public in Nuclear Medicine Department.

1. Introduction

Nuclear medicine uses small amount and low activity of various radioactive sources for nuclear diagnostic and therapy. Radioactive sources that have been used are technetium-99m (^{99m}Tc) [1], Flourine-18 (^{18}F) [2-3] and Iodine-131 (^{131}I) [4] and inserted either via injection or orally into the patient's body. Among these radioactive sources, ^{131}I give highest dose rate due to its natural half-life of 8 days, high mean energy peak of 364 keV and energetic beta minus (β^-) emanations. Beside nuclear imaging, ^{131}I is widely used for radioiodine (RAI) therapy to treat hyperthyroidism and differentiated thyroid cancer [5].

Prior RAI therapy, handling, diluting, and preparing high energy ^{131}I radiopharmaceuticals is always a great challenge [6-7]. During this preparation, radioactive spillage can occur by chances and risk to radioactive contamination. This incident has high potential to give irreversible health-related problems to the patient, staff and public. Meanwhile, patient undergoes RAI therapy will be hospitalized for a few days after taking radioactive ^{131}I and becomes a mobile radiation source [8]. Radioactive ^{131}I that has not been adsorbed by thyroid gland will be leaved patient's body as radioactive waste, mostly via urine and feces [1, 4]. Radioactive wastes were also excreted from sweat, sneeze and puke, which might risk to uncontrollable of environmental radiation contamination. These wastes can affect human health and environment, thus proper and systematic radioactive waste management must be strictly considered [9]. Therefore, it is crucial to recognized efficient decontamination procedures for safe radiation practices. Generally, radioactive spillage can be categorized either major or minor spills, but both identified as an emergency and must be coded [10].

This study aims to propose a low-cost technique for alternative radioactive spillage decontamination in nuclear medicine department by using bamboo activated carbon (BAC) as an adsorbent agent from agriculture waste. Agriculture waste materials have gained attention due to both economic and ecological concern in the last decades and proved significant impact to drive the scientific and industrial communities to look further as alternative organic biomaterials in various applications [11], including for nuclear waste treatment. Bamboo was used due to its vast specific surface area, great porosity and natural antibacterial product that provides super capacity to adsorb and separate septic solutions from the contaminated solvent [12]. In addition, bamboo charcoal has excellent variations of functional groups for radionuclides adsorption such as ^{131}I [13], ^{137}Cs [14-15], heavy metals [16] and Cr (VI) [17].

2. Materials and methods

Bamboo Nano-powders (BNp) was purchased from Zhengzhou Kelin Water Purification Material Corporation Limited, China. Potassium hydroxide (KOH) and hydrochloric acid (HCl) were used in activating the carbon materials. Pure ^{131}I liquid sources and distilled water was supplied by Hospital Universiti Sains Malaysia (HUSM). The instruments used in this study were 1 ml/cc syringe, test tube, lead container, 50 mm micropore filter papers, Geiger-Muller survey meter, dose calibrator (Biodex, Atomlab 500, USA) provided by Hot Laboratory, Department of Nuclear Medicine, Radiotherapy & Oncology, HUSM. Indium Tin Oxide (ITO) glass, Field Emission Scanning Electron Microscopy (FESEM) and Energy Dispersive X-rays (EDX) (FEI QuantaTM 450 FEG, USA) were used at SEM Laboratory, School of Health Sciences, USM for post filtration samples analysis.

2.1. Activation and preparation

Activation process was done by immersed 3 g of BNp into H_2SO_4 solution and H_3PO_4 with a ratio 4:1 [18-21]. The process was followed by addition of $KMnO_4$ into the activation mixture. The mixture was stirred for 3 days and proceed for pouring using 400 ml ice cube with 27 ml of H_2O_2 (30%). The mixture proceeds for centrifuging at 10,000g to get pH 5 and homogenized BAC. The BAC was prepared for 50 mg/ml, 100 mg/ml, 150 mg/ml, 200 mg/ml and 250 mg/ml by manipulating Equation 1:

$$M_1V_1 = M_2V_2 \quad (1)$$

Where,
M_1 = initial concentration of BAC (mg/ml)
V_1 = initial volume of BAC (ml)
M_2 = BAC concentration after dilution (mg/ml)
V_2 = BAC volume after dilution (ml)

2.2. ^{131}I radionuclide adsorption

In this study, 5 ml ^{131}I was diluted with saline water at Hot Laboratory, Department of Nuclear Medicine, Radiotherapy & Oncology, HUSM. The initial activity (A_O) of ^{131}I was determined by using dose calibrator where the reading was 0.7696 MBq. 50 mg/ml, 100 mg/ml, 150 mg/ml, 200 mg/ml, and 250 mg/ml BAC concentrations were mixed with 1 ml of pure ^{131}I as shown in Figure 1(a). Meanwhile, 1 ml of ^{131}I was set as a control group. The radioactive mixtures were left for 15 minutes in lead container to allow initial reactions before filtering.

(a) (b)

Figure 1. (a) 50 mg/ml, 100 mg/ml, 150 mg/ml, 200 mg/ml, and 250 mg/ml BAC concentration was mixed with 1 ml of pure ^{131}I using syringe in fume hood at Hot Laboratory; and (b) Lead container stored the radioactive mixtures during reaction process.

A_O of ^{131}I and its radioactive decay series [$A_O/4$ (day 4), $A_O/2$ (day 8), $A_O3/4$ (day 12) and totally decay (day 16)] were determine from the filter paper, sediment and pure ^{131}I residue after the filtration for kinetic radioactivity study based on Equation 2. Radioactivity rate for sediments and filter paper was compared with control group (natural decay of ^{131}I without BAC mixture) by plotting the decay graph. Geiger-Muller survey meter and electronic pocket dosimeter were used to determine background exposure rate (mSv/hr) for personal radiation protection and safety.

$$A = A_0.e^{-\lambda t} \quad (2)$$

Where,
A = radionuclide activity
A_0 = initial radionuclide activity
λ = decay constant
t = decay time

2.3. Morphology and elemental analysis

Radioactive mixture of 250 mg/ml BAC and 1 ml pure ^{131}I was dropped on ITO glass substrate and left for a few days. The sample was characterized using FESEM and EDX to determine its physical adsorption morphology and elemental analysis at SEM Laboratory. The sample was coated with gold

nanoparticle prior inserted into the vacuum chamber. The sample was disposed according to the radiation disposal procedure recommended by International Atomic Energy Agency (IAEA) after analysis work [22].

3. Results and Discussions

Reaction properties between BAC and ^{131}I mixtures was investigated by physically observed the filtration process. Sediment was formed on the filter papers for each sample after 15 min as shown in Figure 2. Moist was quickly spread for low concentration BAC mixtures and not significantly observed for high concentration. The moist might be due to evaporation of ^{131}I [1, 4, 23-24]. ^{131}I is a type of halogen and has high volatility to vaporize. Meanwhile, less moisture spreading was noticed for high concentration BAC filter paper. This phenomenon might be due to active interactions between BAC and ^{131}I. The amount of ^{131}I trapped in the BAC was elevated and lower vaporization mechanism [23-24]. The observation also shows more volume of sediment has been formed for high BAC concentrations.

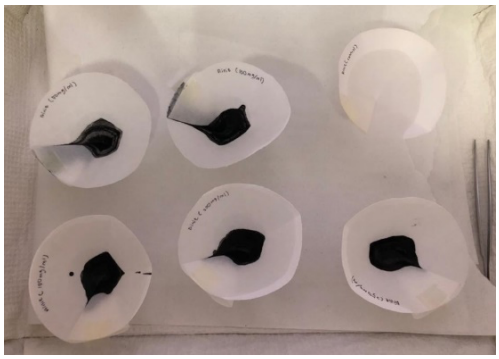

Figure 2. Sediments were formed on each filter papers after 15 min filtration.

3.1. Kinetic study

The radioactivity of ^{131}I residue and sediment after filtration for different concentrations mixtures were plotted in the radioactivity graph for 0, 4, 8, 12 and 16 days as shown in Figure 3(a) and 3(b), respectively. The radioactivity was compared to the control group of pure ^{131}I natural decay (blue line for ^{131}I residue graph and light blue line for sediment).

(a) (b)

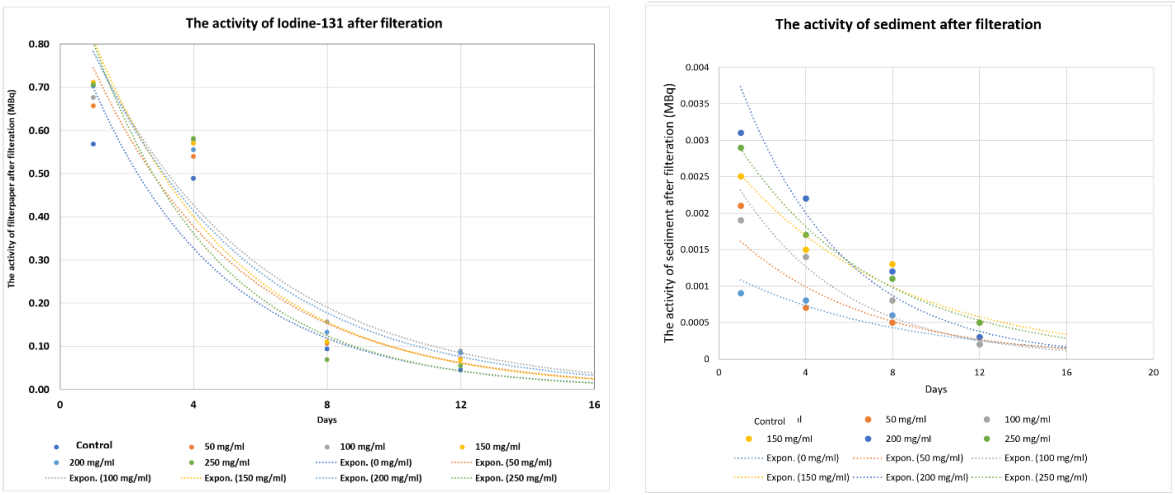

Figure 3. (a) Radioactivity of ^{131}I residue over days after filtration; and (b) Radioactivity of sediment over days after filtration.

Figure 3 (a) shows the radioactivity of ^{131}I residue was exponentially decreased with increasing of BAC concentrations for all decay points. The graph shows that 250 mg/ml BAC has the highest ^{131}I radioactivity followed with 150 mg/ml, 100 mg/ml, 50 mg/ml, and control group on day 1. In addition, the radioactivity was sharply decreased with increasing of BAC concentrations after filtration on days 1st and 8th compared to its natural decay (blue line). The radioactivity loss after filtration was 76.1% (50 mg/ml), 76.3% (100 mg/ml), 83.5% (150 mg/ml), 80.4% (200 mg/ml), 85% (250 mg/ml) compared to 68% (control) due to high agglomeration between BAC and ^{131}I.

Figure 3(b) shows the radioactivity of sediment after filtration on 0, 4th, 8th, 12th and 16th days. It was observed that the radioactivity was accelerated with increasing of BAC concentrations. The graph shows that 200 mg/ml BAC has the highest radioactivity followed with 250 mg/ml, 150 mg/ml, 50 mg/ml, 100 mg/ml and control group on day 1 (light blue line). The radioactivity of ^{131}I was sharply decreased with increasing of BAC concentrations after the filtration within day 1st until day 8th. Meanwhile, the radioactivity of ^{131}I for 200 mg/ml concentration has slightly decreased than its natural activity on day 12th. The radioactivity of sediments was reduced by 0.2% for 50 mg/ml, 100 mg/ml, 150 mg/ml, 0.01% for 200 mg/ml, 0.3% for 250 mg/ml and 0.09% for control group (naturally decay without BAC).

Initial adsorption of ^{131}I was very quick followed by gradual adsorption which approaching the equilibrium within day 12th. The radioactivity of BAC mixtures concentration was always higher than control group due to solidification that agglomerate the radioactive ^{131}I on the filter paper [4]. Generally, radioactivity of the filter paper for all different concentrations were higher than the trendline on the day 4th as shown in Figure 3 (b), where higher concentrations of BAC mixture (200 mg/ml and 250 mg/ml) led to obtain highest radioactivity readings. Meanwhile, radioactivity of control group was less at all points due to no agglomeration occurred to trapped ^{131}I on the filter paper. However, the adsorption was believed to only accelerated the radioactivity loss by chemical molecular interactions [25], without changes the identity of original radionuclide.

3.2. Sediment characterization

Figure 4 (a) and 4(b) show the pores with different sizes and shapes existed on the external surface of 250 mg/ml of BAC:^{131}I mixture with different magnification. The sample surface becomes coarser and might be due to the deposition of ^{131}I radionuclide and other elements during the interaction mechanisms [23-24]. The micrograph shows there was adsorption mechanism occurred where non-uniform particles were trapped on the surface structures. The adsorption was contributed by various functional groups (carboxyl, carbonyl, and hydroxyl groups) existed on the BAC structures [13]. There were many agglomerated non-uniform particles developed on the carbon surface and probably formed during rapid interactions between BAC and ^{131}I after 15-minute of filtration.

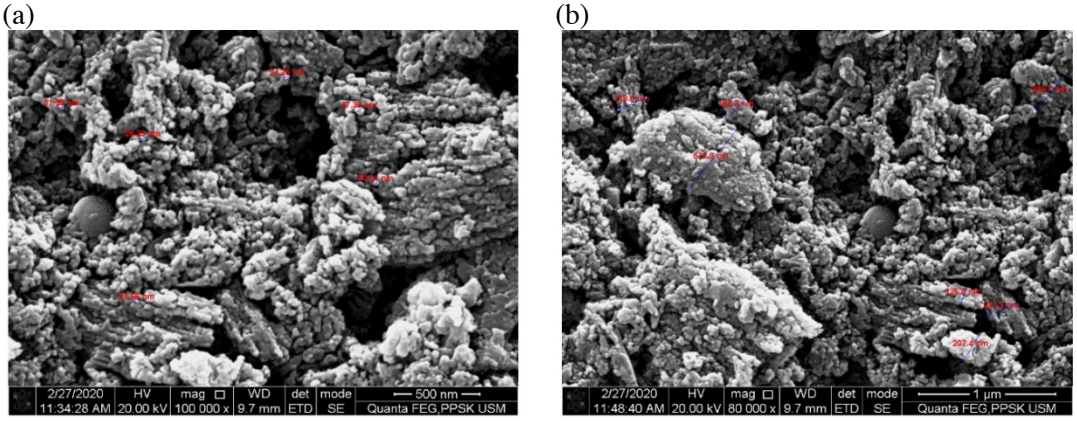

Figure 4. Micrograph of 250 mg/ml BAC:^{131}I mixture shows some non-uniform particles agglomerated on the different BAC structures for (a) 100 000x and (b) 80 000x magnifications.

Figure 5 represents the elemental composition from the 250 mg/ml BAC:^{131}I dried mixture. The graph presented a small weightage of Na element which was part of ^{131}I compound (Na_2I). This element proved the agglomeration of ^{131}I onto BAC surface (4). However, ^{131}I element was not detected and might be physically loss with no significant considerable amounts has been left due to total decay after day 16th. There were also other elements detected such as Carbon (C), Oxygen (O), Aluminium (Al), Silicon (Si), Chlorine (Cl) and Calcium (Ca), probably originated from chemicals that has been used during activating process. Carbon (89.71%) and oxygen (9.77%) were the highest elements present in the sample due to natural carbonaceous and oxygenase compounds originated from BAC.

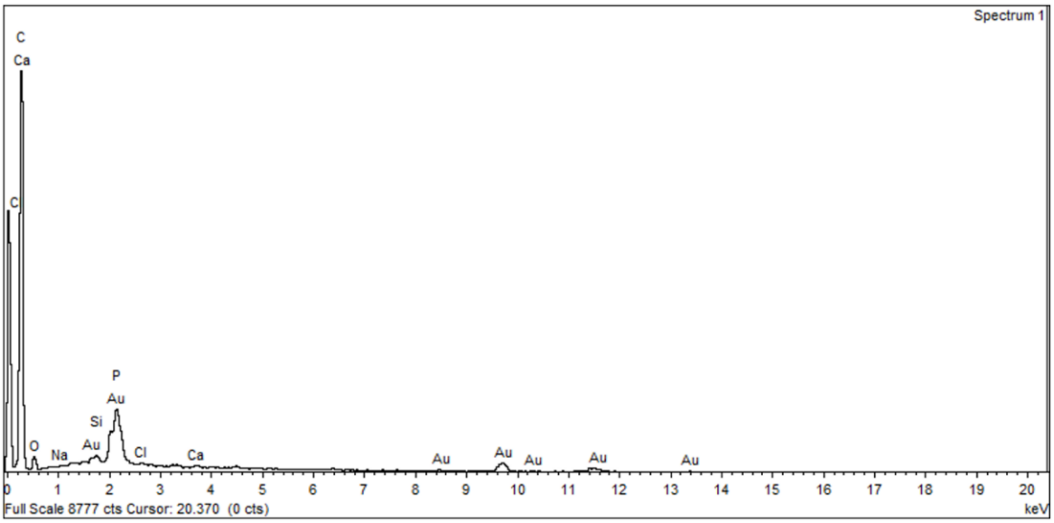

Figure 5. Elemental analysis of 250 mg/ml BAC:^{131}I sediment shows the present of Na element which was part of ^{131}I compound.

4. Conclusion

The adsorption mechanisms show complicated interactions between BAC and ^{131}I. The results show the activity of sediment was increased for initial filtration due to active agglomeration with increasing of BAC concentrations. 250 mg/ml BAC concentration able to accelerate the radioactivity loss by 85% compared to the control group which was 68%. FESEM image proved there were porous structures on the BAC and contributed to attract foreign particles via agglomeration interactions. EDX revealed the present of Na on the BAC surface which might be part of ^{131}I compound. This study revealed that BAC performed different capabilities as an alternative adsorbent material under different experimental conditions and has high potential for sustainable radionuclide decontamination agents especially for RAI therapy in ensuring healthy environment for staff, patients and public.

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