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**Effect of impregnating materials in activated carbon on Iodine -131 ( 131 I) removal efficiency**  
  
[Maghsoud Gourani](http://www.rpe.org.in/searchresult.asp?search=&author=Maghsoud+Gourani&journal=Y&but_search=Search&entries=10&pg=1&s=0)**1,**[Asghar Sadighzadeh](http://www.rpe.org.in/searchresult.asp?search=&author=Asghar+Sadighzadeh&journal=Y&but_search=Search&entries=10&pg=1&s=0" \t "_blank)**2,**[Farhang Mizani](http://www.rpe.org.in/searchresult.asp?search=&author=Farhang+Mizani&journal=Y&but_search=Search&entries=10&pg=1&s=0" \t "_blank)**3**  
1 Safety and Environmental Lab, Nuclear Science and Technology Institute, Atomic Energy Organization of Iran, Tehran; Department of Chemistry, Faculty of Science, Payam Noor University, Tehran, Iran  
2 Safety and Environmental Lab, Nuclear Science and Technology Institute, Atomic Energy Organization of Iran, Tehran, Iran  
3 Department of Chemistry, Faculty of Science, Payam Noor University, Tehran, Iran

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**Correspondence Address**:  
Asghar Sadighzadeh  
Nuclear Science and Technology Institute, Tehran   
Iran  
[Login to access the Email id](http://www.rpe.org.in/login.asp?rd=article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani)

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| **Abstract** |  |  |

Activated carbon (AC) is widely used in various industries as an adsorbent material. It is used in gas cleaning industries, because of its low cost and high efficiency for removal of pollutants from effluents. It can be produced from a wide range of agricultural activities as by-products. There are different methods for producing the AC. The most common methods are physical and chemical activation that includes heat treatment, amination and impregnation. In this study, the effect of three impregnates, i.e., NaOH, KI and ZnCl 2 on the quality of AC for 131 I removal, was investigated. Our results show that the sodium hydroxide impregnated AC is more effective for 131 I removal. Also, the quality of the impregnated AC was decreased in the case of impregnants material percentage exceeding 2%wt.

**Keywords: Activated carbon, efficiency, I-131, impregnation, removal**

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| **Introduction** |  | [Top](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani#top) |

Activated carbon (AC) is used for a wide range of applications, such as gases separation, solvent recovery, organic pollutants removal from drinking water, catalyst support and various industrial sectors such as food, pharmaceutical and chemical industries. In the nuclear industry, it is used to adsorb radio-iodines from the gaseous effluents. It is the most widely used adsorbent material because of its large adsorption capacity and low cost. [[1]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref1)   
  
Radioactive 131 I is one of the most environmentally hazardous radionuclides owing to its potential for high uptake by human body, especially by the thyroid gland. [[2]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref2) Radioactive iodine is released in the environment from accidents in nuclear reactors or from leakages/incidents in 131I production plants when it could not be absorbed by alkali solution in sublimation method. Due to the existence of a variety of chemical species of iodine in such releases, the efficient removal of radioactive iodine from gaseous aerosols is a complex problem. [[2]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani#ref2)  
  
There are several methods of gaseous radioactive iodine pollution treatment, such as AC and wet scrubber. AC method is more reliable. It is commonly used in the radiopharmaceutical industry. AC has the strongest adsorption properties because of its large surface area provided by of the pores. It is a black, solid substance that is commercially available in different shapes such as granular or powder and is extremely porous with a very large specific surface area. Its surface area can reach up to 1,000 m 2 /g. For example, 5 g of AC can have the surface area of a football field. [[3]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref3)  
  
Activated carbon mainly consists of carbon (87-97%) and other elements such as hydrogen, oxygen, sulfur and nitrogen. [[4]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref4) The word activation refers to the development of the adsorption properties of carbon. Raw materials such as coal and peanut shells have different adsorption capacities, and this property can greatly enhanced by activation process. There are two processing methods for production of AC including physical and chemical activation. Surface area and pore volume are the two main parameters that decide the performance of the AC. The size and volume of pores are important for maximizing adsorption. The International Union of Pure and Applied Chemistry define micropore, mesopore and macropore as follows: [[5]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref5)

* Micropore, pore diameter <2 nm
* Mesopore, pore diameter in the range of 2-50 nm; and
* Macropore, pore diameter >50 nm.

The macrospores are used as the entrance to the AC, the mesopores for transportation and the microspores for adsorption. The porosity of AC can be measured using iodine number. [[4]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani#ref4) The iodine number (mg/g) is a measure of the micropore content of the AC by adsorption of iodine from solution when the iodine concentration in the residual filtrate is 0.02 normal. [[6]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref6)  
  
According to the aforementioned descriptions, the impregnated AC is known as one of the best ways to 131 I adsorption that is received significant attention of researchers all over the world.  
  
The removal of radio-iodine has generally been performed using AC, silica, alumina, and polymer resin. Also impregnants such as KI, triethylenediamine (TEDA) and silver have been widely used to improve the removal efficiency of AC. There has been some concern regarding the use of TEDA-impregnated AC as an adsorbent for radio-iodine removal. [[7]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref7),[[8]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref8)  
  
**Preparation of activated charcoal**  
  
Activated carbons were prepared by different series of carbon dioxide and steam activation from walnut shells for their optimal use as radioactive methyl iodide adsorbents in nuclear plants. In order to increase their methyl iodide affinity, the effect of TEDA impregnation was studied at 5 and 10%wt. The results obtained indicated that in both cases the adsorption efficiency is markedly improved by the addition of impregnants, which allows the adsorbate uptake to occur not only by physical adsorption, via nonspecific interactions (as in nonimpregnated carbons) but also by the specific interaction of TEDA with radioactive methyl iodide. Methyl iodide retention efficiencies up to 98.1% were achieved. [[9]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref9) Investigations by Srevastava showed that AC, impregnated with KI/KOH, removes radio-iodine by isotope exchange as follow:[[10]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref10)  
  
CH 3131 I + K 127 I→CH 3 127 I + K 131 I  
  
When volatile radio-iodine 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. [[11]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref11) The effect of ageing on the performance of KI-impregnated charcoals has been examined, and some observations have been made on the mechanism of the reaction between methyl iodide and iodide ion on a carbon surface. [[12]](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani" \l "ref12)  
  
In this work, KI, NaOH and ZnCl 2 were used to impregnate AC with the aim of improving its gaseous radio-iodine removal. Also, the impact of impregnant concentration on the pore size and the mechanism of Iodine removable increasing were investigated. The source of radio-iodine was the leakage from the hot cell of 131 I production into exhaust air. In accordance with the Srevastava study, also our investigation on KI impregnation show that absorption occurs in both surface and pores of AC.  
  
**Experimental**  
  
The granular AC is used in this study in order to investigate the impact of impregnating material on 131 I removal efficiency, is prepared by nut shells. The effective surface area of it checked by BET method was 950-1,000 m 2 /g and the dimension of granular was 0.7-1 mm. The adsorbent material used for improving the efficiency of GAC was KI, ZnCl 2 and NaOH solutions.  
  
In order to impregnate fresh AC of 40 g, it was rinsed three times in deionized water for removing the impurities, and then it was dehydrated at 110°C. The prepared AC submerged in different %wt solution prepared by one of the adsorbent material quoted above. The submerging time was 3 h. Then the sample was dehydrated at 110°C for 5 h. Surface area checked by Brunauer-Emmett-Teller (BET) method for different %wt[[Table 1]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_t6.jpg" \t "_blank).

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| http://www.rpe.org.in/articles/2014/37/3/images/RadiatProtEnviron_2014_37_3_179_154882_t6.jpg | Table 1: Surface area of different %wt (m2/g)    [**Click here to view**](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_t6.jpg) |

Results of BET method shows, %wt and surface area, are inversely proportional, when the %wt is more than 2%. Sudden decrease of surface area more than 2%wt impregnation represents the correct choice impregnate as 2% by wt.  
  
[[Figure 1]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f1.jpg) indicates that, surface area changes of <2%wt impregnation is too slow and then suddenly rises. Therefore, the optimal of 2%wt is selected. Thus, three samples of impregnated GAC prepared.

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| http://www.rpe.org.in/articles/2014/37/3/images/RadiatProtEnviron_2014_37_3_179_154882_f1.jpg | Figure 1: Surface area of different %wt  [**Click here to view**](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f1.jpg) |

The obtained samples were kept in a sealed container to prevent adsorption of moisture and impurities from the air. The qualities of prepared samples are tested by 131 I radionuclide gas. These samples are set in outlet of 131 I production system where the discharging gas passed to nuclear installation and chimney. The test set up includes a column of granular AC bed (GACB) with 5 and 15 cm of diameters and length respectively, followed by a Standard Activated Carbon Filter Paper (SACFP) produced by Schleicher and Schuell Co. The grade, diameter and radio-iodine removal efficiency of SACFP, were 509, 60 mm and 99.8%, respectively. A vacuum pump (Push Co. S50006F, 6 m 3 /h) was used to provide the polluted air flux through the set-up. The test air flux was fixed at 10 L/min. The amount of AC in each experiment was 40 g. The efficiency of the granular bed was measured by two different radio-iodine concentrations. The iodine concentration of the first series was practically two times more than the second series.  
  
The efficiency of GACB was calculated by relation (1):  
  
*E* = [GC GACB /(GC GACB + GC SACFP )] ×100 (1)  
  
Where, GC GACB and GC SACFP are gamma count of GACB (Bq/min) and SACFP (Bq/min) respectively. A gamma counter with Sodium iodide detector was served to measure the activity of GACB and SACFP.

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| **Results and discussions** |  | [Top](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani#top) |

In this research, the 131 I removal efficiency of three impregnated AC sample with KI, ZnCl 2 and NaOH have been investigated. The iodine removal efficiency was determined by gamma counting of the tested sample and filter. The experimental gamma counting result was shown in[[Table 2]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_t7.jpg" \t "_blank).

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| http://www.rpe.org.in/articles/2014/37/3/images/RadiatProtEnviron_2014_37_3_179_154882_t7.jpg | Table 2: Gamma count of GACB and SACFP    [**Click here to view**](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_t7.jpg) |

The efficiency of granular beds is computed using the relation 1. The results of the AC efficiency for removal of 131 I gas pollutant for samples inclusive without impregnation and with 2%wt impregnant KI, ZnCl 2 and NaOH are 54%, 59%, 70% and 90% in first tests and 56%, 74%, 79% and 99.65% in second test, respectively. The disparity between removal efficiencies of first and second series may be explained by the differences between concentrations of radio-iodine in two series of experiments [[Figure 2]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f2.jpg).

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| http://www.rpe.org.in/articles/2014/37/3/images/RadiatProtEnviron_2014_37_3_179_154882_f2.jpg | Figure 2: Efficiencies of samples with two different iodine-131 concentrations  [**Click here to view**](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f2.jpg) |

Record numbers of test series two are the average of results obtained and are summarized here.  
  
Pretests with different percentages of impregnation showed excellent absorption impregnation is 2% because impregnation in small amount due to lack of adequate coverage holes, and impregnation with higher percentages results in the fully occupied space inside the hole, and absorption is strongly reduced. The following graph represents the absorption efficiency is impregnated with different percentages of NaOH as impregnant [[Figure 3]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f3.jpg).

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| http://www.rpe.org.in/articles/2014/37/3/images/RadiatProtEnviron_2014_37_3_179_154882_f3.jpg | Figure 3: Efficiency of NaOH impregnant by different %wt  [**Click here to view**](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f3.jpg) |

The impregnated AC radio-iodine removal is the sum of absorption and adsorption processes. 131 I absorption relation for the AC specimen which impregnated by KI, ZnCl 2 and NaOH are as follows:  
  
KI + 131 I 2→ K 131 I 3 (1)  
  
ZnCl 2 + 131 I 2 (g)→ Zn 131 I 2 (Cl 2 ) (2)  
  
131 I 2 + 2NaOH → Na 131 I + Na 131 IO + H 2 O (3)  
  
The moisture created by the air and reaction (3) will react with NaOH, and additional chemical absorption will occur according to the following reaction. Chemical absorption of iodine by the two processes (reactions 3, 5), increase the total efficiency of NaOH impregnated AC.  
  
NaOH (S) + H 2 O (l)→ Na + + OH− + H 2 O (4) × 2Na + + 131 I2→ 2Na 131 I (5)  
  
As seen in [[Figure 2]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f2.jpg), total absorption of NaOH impregnated AC is higher than KI and ZnCl2 impregnated AC. It is in the order of 20-25%. Therefore, further studies about concentration effect of impregnant are performed by only for NaOH impregnated specimen.  
  
[[Figure 4]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f4.jpg) shows the pores of AC without any additive. According to [[Figure 4]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f4.jpg), there are numerous pores in the structure of AC. AC with its characteristics could be an applicable adsorbent with high ability for trapping the specific pollutants from the air if these pores are impregnated with appropriate impregnants.  
  
[[Figure 5]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f5.jpg) shows the morphology of AC before (raw AC), and after impregnation process (2%wt NaOH, 4%wt NaOH and 10%wt NaOH) as mentioned in the experimental section.

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| http://www.rpe.org.in/articles/2014/37/3/images/RadiatProtEnviron_2014_37_3_179_154882_f4.jpg | Figure 4: Activated carbon (×43) before impregnation  [**Click here to view**](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f4.jpg) |

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| http://www.rpe.org.in/articles/2014/37/3/images/RadiatProtEnviron_2014_37_3_179_154882_f5.jpg | Figure 5: Scanning electron microscopy none and impregnated activated carbon (×2,200)  [**Click here to view**](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f5.jpg) |

[[Figure 5]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f5.jpg)a shows the large pores of AC without any additive. It should be noted that, more than 50% of 131 I was trapped in micro, meso and macropores. [[Figure 5]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f5.jpg)b indicated that how 2% NaOH solutions bonded by surface but the holes are still visible. Thus, both type of trapping including absorption and adsorption should be considered. [[Figure 5]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f5.jpg)c shows the effects of 4% NaOH impregnant solution on the AC structure. The pores are quasi-filled by impregnant, and little shallow holes are visible. Thus, the adsorption effect practically will not take place.  
  
In [[Figure 5]](http://www.rpe.org.in/viewimage.asp?img=RadiatProtEnviron_2014_37_3_179_154882_f5.jpg)d, the AC specimen impregnated with 10% NaOH is presented. As it is observed, pores are not visible. In effect, the pores are totally fulfilled by impregnant. Consequently, the physical adsorption will not take place.

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| **Conclusions** |  | [Top](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani#top) |

The effective surface area of AC prepared in this study was ranging from 950 to 1000 m 2 /g. The Iodine absorption efficiency of the AC sample was equal to 54%. Among the three tested impregnating materials (KI, ZnCl2and NaOH), NaOH showed the highest Iodine adsorption efficiency. The radio-iodine removal efficiency of NaOH impregnant sample was determined as 99.65%. This was equivalent to practically 46% of the rise in gas removal efficiency using NaOH as impregnating material.  
  
In this paper, it is shown that the impregnant concentration exceeding 2% causes the pores plugging. This means radioiodine adsorption efficiency will be decreased by increasing impregnant concentration. This can be explained by relative saturation of AC granular bed.

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| **References** |  | [Top](http://www.rpe.org.in/article.asp?issn=0972-0464;year=2014;volume=37;issue=3;spage=179;epage=183;aulast=Gourani#top) |

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