Removal of cesium through adsorption from aqueous solutions: a systematic review

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ABSTRACT

Cesium radioactive isotopes (^{134}Cs and ^{137}Cs) are dangerous to human health due to their long half-life and high solubility in water. Nuclear experiments, wars, and nuclear plant accidents have been the main sources of Cs release into the environment. In recent years, several methods have been introduced for the elimination of Cs radioactive isotopes from contaminated water. This study provides an overview of the available published articles (2008–2016) regarding the remediation of waters polluted by Cs isotopes. The maximum adsorption capacity (MAC) of Cs isotopes corresponded to natural chabazite (273.24 mg Cs/g adsorbent), hollow Prussian blue (PB) nanoparticles (262 mg Cs/g adsorbent), and Prussian blue implemented non-woven fabric (260 mg Cs/g adsorbent). Using natural chabazite and PB adsorbents, along with another adsorbent (PB graphene oxide hydrogel), Cs radioactive isotopes can be effectively removed from the aqueous solutions.

Keywords: Cesium, Isotopes, Sorption, Biosorbents, Drinking Water, Radioactive Material

Introduction

In recent decades, radioactive material toxicity has attracted significant attention worldwide. In the recent earthquake in Japan, on March 11, 2011, large amounts of Cesium-134 (^{134}Cs) and Cesium-137 (^{137}Cs) radioactive materials (630,000–770,000 TBq) were discharged into the water, air, and soil.[1,2] ^{137}Cs is the most abundant radionuclide and has devastating effects on the environment due to its high water solubility and long half-life.[3]

Furthermore, ^{137}Cs is a source of beta emission; therefore, it is called "heat generator".[4] The chemical behavior of ^{137}Cs is similar to sodium and potassium; hence, it can be deposited in the soft tissues of aquatic and terrestrial organisms.[5] In the human body, ^{137}Cs can cause impairment of the reproductive system, liver, kidneys, and central nervous system, as well as cause behavioral disorders.[6] Several techniques have been used to remove ^{137}Cs from aqueous solutions including precipitation,[7] extraction,[8] ion exchange,[9] bioaccumulatio,[10] and adsorption process.[11,12,13,14]

When we use the conventional water treatment methods, such as coagulation–sedimentation and sand filtration (10%–40% efficiency), removal of ^{137}Cs from the

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contaminated water is difficult.[15] Significant efforts have been made to remove ^{137}Cs from the aqueous solution.[15] Adsorbents including zeolites, crystalline silicotitanate, aluminum molybdophosphate, metal-ferricyanide, ferricyanide, and ammonium 12-molybdophosphate (AMP) have been tested for the removal of ^{137}Cs from waters.[16] Natural absorbents have low efficiency in removing ^{137}Cs from water, although, expensive synthetic absorbents show high efficiency.[17] Prussian blue (PB)-based adsorbents are efficient ^{137}Cs scavengers because the $Fe_7(CN)_{18}$ matrix has a crystal cage size similar to the hydration radius of Cs ions.[17,18] The efficiency of adsorbents in removing Cs depends on the chemical structure of the adsorbents, pH, temperature, and so on.[19] The main objective of this study was to provide information on the current innovations about the removal of ^{137}Cs from aqueous solutions using adsorbent process.

Materials and Methods
Literature search
A The search of the available studies and their analysis was carried out by three investigators (Ya.F, Me.A, and Ya.Z). Almost all the studies regarding the Cs removal from water solutions werescreened ininternational and national databases. The international databases searched were Web of Science, PubMed, Scopus, ScienceDirect, and Google Scholar.

The keywords used were cesium, cesium, aqueous solutions, water, wastewater and its removal, treatment, radioactive, and radioactive liquid waste. The data published from January 2008 to October 2016in the English language databases was collected. We also searched the bibliographies of retrieved articles for additional references. The literature search was based on PRISMA guideline (Fig. 1).[20]

Inclusion and exclusion criteria
Original articles were included if they satisfied all the following criteria: (1) is an original article; (2) is a laboratory study about the removal of Cs from aqueous solutions by the adsorption process; (3) is written in the English

language; (4) was published online between January 2008 to October 2016; (5) full-text article is available.

Data extraction and definitions
The following variables were extracted from the included studies: adsorbent and maximum adsorption capacity (Table 1). Inconsistencies between the reviewers were discussed to obtain consensus through a focus group. The references of the selected articles were checked to identify all articles not found in the databases.

Results and Discution
Four hundred ninety-one articles were reviewed separately by three investigators. Forty-two articles (73 studies) were selected for the final review. The release of radionuclides such as Cs, radium isotopes ([226]Ra and [228]Ra), radon 222 ([222]Rn), and thoron (Tn) into the environment, particularly into drinking water sources, can threaten human health.[21,22,23,24] The maximum acceptable level of uranium (U) in drinking water is 30 μg/L, [228]Ra and [226]Ra combined is 5 PCi/L, the activity of alpha particles is 15 PCi/L, and that of beta particles and photons is 4 mrem/year.[25] With the release of beta particles, radio Cs is converted to [134]Cs (half-life of 2 years) and [137]Cs (half-life of 2.5 years).[25]
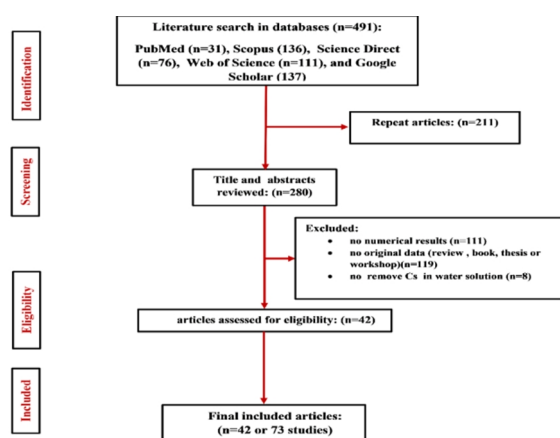

Fig. 1. Selection process of articles on the remove Cs from aqueous solutions by adsorption process

After 10 days of the Fukushima nuclear accident, Tokyo's drinking water was contaminated with Iodine-131(^{131}I), and two

years later, traces of Cs radionuclides were still detectable in many of the Japanese cities.[26] Even at low levels, Cs radionuclides in drinking water could be harmful because they can accumulate in the human body.[27,28] In the conventional water treatment plants, raw water that entered into a grain pond after its passage through coarse and fine dirt was stuck in the first place. Then, coagulants such as alum, ferric chloride, and ferric sulfate were added to the water. The formed flocs then settled in the sedimentation basin. The floc deposits were then collected from the bottom of the pond. Finally, the water was passed through slow or fast sand filters, so the tiny particles could be removed. Passing of water through sand filters made it clear. The water was then disinfected using chlorination units before it entered the water distribution system.[29] In the study by Morton et al. the average removal of Cs by the Jar test was 5%.[30] Gäfvert et al. reported that the removal of Cs was low in large wastewater treatment plants, in which alum and ferric sulfate were used as coagulants (110000 m³/d). They reported that the low turbidity in the Lake Bolmen water source was the main cause of decreased efficiency in Cs removal because the adsorption of Cs on the particles was reduced.[31] Goossens et al. suggested that adding alum sulfate as a coagulant, together with activated carbon and activated silica, to the sand filters removed 31% and 25% of the coagulation, respectively, in the filtration units.[32] Baeza et al. used ferric sulfate and ferric chloride as natural water coagulants and studied the radionuclide removal efficiency in the coagulation and flocculation process. The results showed that the efficiency of Cs removal was low (5.2%). The reason for the low efficiency of Cs removal was that the Cs was in a water-soluble form; therefore, coagulation was ineffective in the removal of Cs.[33] Kosaka et al. investigated the concentrations of ^{131}I, ^{134}I, ^{134}Cs, and ^{137}Cs in five water treatment plants in Japan, and concluded that ^{131}I treatment was not successful by the coagulation–sedimentation process, but 30%–40% of it was removed by adsorption on activated carbon. Radioactive Cs attached to the particles was

effectively removed by the coagulation and flocculation process, but the Cs ions (solution form) were not eliminated.[34] Brown et al. suggested that if the Cs was in a particle-bonded form, it was removed by the coagulation and flocculation process, in the range of 10%–40%, but if it was in a solution form, minimum removal efficiency was achieved.[35]

Cs adsorption processes

Different biosorbent shave been used for the Cs removal including hollow PB nanoparticles, bentonite, mesoporous hybrid adsorbent, vermiculite modification by ethylamine, montmorillonite-iron oxide composite, and NaOH-treated moss (Table 1).[14] Since the adsorption process is more efficient and less expensive for Cs removal, extensive studies on Cs removal by a variety of surface absorbents have been carried out worldwide. These studies examined the adsorption at low cost.[14] Many extracellular polymeric substances are often present in a cell mass, which increase the affinity of Cs to absorb.[36] In the study by Borai et al., it was shown that natural mordenite had more adsorption capacity for ^{134}Cs from liquid waste than natural clinoptilolite (NaNCl), natural chabazite (NaNCh), and synthetic mordenite (NaSM) (Table 1).[37]

The radius of a bare Cs ion is 3.40 Å and is, therefore, easily accommodated by the large (smallest diameter 6.5 Å) 12-ring cavities in their mordenitestructure.[37,83] Brown et al. showed that the use of activated carbon and zeolite removes 0%–10% and 40%–70% of Cs from water, respectively. The turbidity removal capacity increases with an increase in turbidity.[35] Liang et al. suggest that the adsorption of Cs and strontium (Sr) from water by mordenite is mostly done by the mechanism of ion exchange.[84] Ion exchange also increases radionuclide adsorption.[84] Dyer et al. showed that use of pillared inter-layer clays (with high-level and high-ion exchange capacity) increased the affinity of Cs to absorption in comparison with calcium, potassium, ammonium, sodium, and magnesium.[85]

Table 1. Recent studies for Cs adsorbents considering maximum adsorption

Adsorbent	Maximum adsorption capacity (mg Cs/g adsorbent)	Ref
Natural chabazite	273.24	37
Hollow prussian blue nanoparticles	262	38
Pb implemented non-woven fabric	260	39
Natural mordenite	254.76	37
Prussian blue (pb) granules	241	39
Novel magnetic zeolite nanocomposite	229.3	40
Synthetic mordenite	220.44	37
Phosphoric acid activated montmorillonite	208	41
Bentonite	176.09	42
Natural clinoptilolite	167.64	37
Prussian blue caged in spongiform 167	167	43
Prussian blue graphene oxide	164.5	13
Kznhcf-pan	161	44
Mucilaginous seeds of ocimumbasilicum	160	45
Prussian blue alone 158	158	43
Pan-knicf	157.7	46
Cs-g-bentonite	153.65	47
Prussian blue caged in alginate/calcium beads	142.85	48
Reinforced with carbon nanotubes	131.57	48
Prussian blue caged in alginate/calcium beads	120.9	49
Clinoptilolite	118.00	50
Titanium phosphates	114.9	51
Ferrite	97.63	52
Mesoporous hybrid adsorbent (mha)	96	17
Pb-coated mnpnano-sorbents	91.8	53
Ammonium molybdophosphate–calcium alginate composite adsorbent	87.72	54
Copper ferrocyanide is fixed organic silica	86.28	52
Hybrid adsorbent	81.3	55
Ammonium molybdophosphate–Polyacrylonitrile	78.17	42
Vermiculite modification by	77.70	56
Ethylamine	72.6	44
Conjugate adsorbent	68.49	51
Potassium metal hexacyanoferrate - pan	56.92	42
Magnetite	56.00	57
Ethylamine	55.32	58
Prussian blue (pb) impregnated in alginate gel (Ag)	53.2	8
Coconut shell activated carbon	53.00	57
Prussian blue analogues CO3[FE(CN)6]2 H2O (COFC)	52.6	59
Prussian blue (pb)	50.16	8
Montmorillonite-iron oxide composite (Mioc)	43.96	47
CO3[FE(CN)6]2.H2O (COFC)	39.6	60
Cnts	37.8	61
Taiwan laterite	37.63	62
Synthetic a-x zeolite blend	29.33	63
Sm-amp20	29.3	38
Taiwan laterite	37.63	62
Synthetic a-x zeolite blend	29.33	63
Taiwan laterite	37.63	62
Synthetic a-x zeolite blend	29.33	63
Sm-amp20	29.3	38
Copper ferrocyanide (cufc)	17.16	8
Commercial pb particles	17.1	64
Cofc-silica-py	17	65
Copper ferrocyanide functionalized mesoporous silica) (seawater, ph 7.7)	16.58	66
Naoh treated moss	16.2	67
Nickel hexacyanoferrate	15.15	68
Prussian-blue-modified magnetite (pb–fe3o4)	12.75	69
Clinoptilolite	12.5	64
Oxidized multiwall carbon nanotubes	10.56	70
Iron(iii) hexacyanoferrate (ii) (insoluble prussian blue)	10.56	71
Crushed granite	10.1	70
Carbonized rice hull	7.3	72
Brewery's waste biomass	6.68	73
Copper hexacyanoferrate–polyacrylonitrile composite (chcf–pan)	6	74
Sericite	5.5	75
Moos	5.43	75
Copper ferrocyanide	5.34	75
Iron pillared layered montmorillonite (fe-pilm)	5.28	59
Their mixture (fe-pilm;α-feooh)	4.94	76
Cofc/glass-py	4.31	77
Nickel (ii) hexacyanoferrate (iii) residue-walnut shell (niiihcfiii-ws)	4.04	78
Ainoussera	4	79
Zirconium tungstate	3.96	71
Kaolin	3.93	80
Sawdust (beech)	3.05	81
Pre-treated arca shell biomass	1.32	82
Goethite (α-feooh)	0.1	76

Li *et al.* used vermiculite for adsorption of sodium from the water. Cs removal efficiency of 98% was achieved in 5 hours (1.5 µg/g).[86] The results of our study also showed that the presence of sodium, calcium, and ammonium ions reduced the uptake of Cs radionuclides. The first study of Kim *et al.* suggested that Yesan clay and zeolite removed 1.865 and 9.055 µg/g of Cs from the water, respectively.[87] In the second study of Kim *et al.*, sericite was used to remove Cs from water, and the maximum value of adsorption was 6.68 µg/g.[73] Ding *et al.* used walnut shell integrated with nickel hexacyanoferrate to absorb Cs from the aqueous solution. The amount of Cs and Nickel hexacyanoferrate integrated Cs (NiHCF-WS) adsorption was 0.1 mg/g and 0.5 mg/g, respectively. HCF-WS could absorb Cs effectively through a spontaneous

endothermic process.[76]

Ararem et al. showed that montmorillonite-iron oxide composite had a maximum Cs adsorption of 52.6 mg/g in aqueous solution.[59] Long *et al.* compared Cs adsorption by ethylamine-modified montmorillonite (Ethyl-MT) and calcium-saturated montmorillonite (Ca-Mt). The results showed that Ethyl-MT had a greater Cs removal efficiency because of a higher absorption capacity and also greater surface.[88] Bayulken *et al.* investigated Cs adsorption by Turkish clay (bentonite, zeolite, sepiolite, and kaolinite). The results showed that bentonite and zeolite had greater adsorption capacity than sepiolite and kaolinite.[89] Du *et al.* used the spherical PAN-based potassium nickel hexacyanoferrate for Cs adsorption. The results showed that Cs had a greater binding affinity than sodium, magnesium, calcium, potassium, and ammonium ions.[46] Chitrakar *et al.* showed that manganese oxide layered in pH = 2–4 and pH = 10 removed 172 mg/g and 132 mg/g of Cs from the water, respectively.[90] Dahiya *et al.* studied the adsorption of lead, copper, cobalt, and Cs from water and concluded that the maximum level of absorption for Cs was 3.93 ± 0.11 mg/g.[80] During the first study by Krishna *et al., Funariahygrometrica* biosorbentswere used to remove ^{137}Cs and ^{90}Sr. The study showed that Cs was removed from the aqueous solution in the capacity of 38 mg/g. The –COO group is also responsible for the radionuclide absorption. In the second study by Krishna *et al., Funariahygrometrica* was mixed with polysilicates and used to remove ^{137}Cs and ^{90}Sr.[65] At pH 5–10 and 30-minute contact time, the absorption level was 15 mg/g. A number of studies showed undesirable adsorption. For example, during a study on Cs removal by coconut shell–activated carbon, Caccin *et al.* showed that activated carbon has a low absorption capacity.[58] Suzuki *et al.* used vermiculite for Cs absorption, which did not have adequate absorption in artificial seawater.[91] Long *et al.* increased the vermiculite's specific surface area and porosity for Cs removal by ethylamine. The specific surface area increased from 4.35 to 15.59 m^2/g. Also, the average hole diameter decreased from

5.34 to 6.8 nm. The rate of increase of Cs aqueous solution absorption rose from 56.92 to 78.17 mg/g after the vermiculite was modified. The structure of the holes and the specific surface had an important role in Cs adsorption. The presence of cations ($Ca^{2+} > K^+ > Na^+$) and organic molecules with low molecular weight prevented the adsorption of Cs.[42] Based on the above studies, use of absorbents, except vermiculite, did not have a good efficiency in Cs removal. Alkaline compounds could also compete with Cs in water adsorption. Among the Cs special absorbents, competition between Cs and alkaline compounds was less, and Cs was absorbed on a priority. Therefore, Cs special absorbents such as old dye and PB were used.

Removal of Cesium by Prussian blue

PB was demonstrated as an efficient Cs scavenger because $Fe_7(CN)_{18}$ had a crystal cage size similar to the hydration radius of a Cs ion. Therefore, a separate section is devoted to PB. PB is a lattice structure with eight molecules of water. It was used for adsorption of Cs from the victims' bodies after the Chernobyl incident in 1987. On account of the high affinity of Cs to PB, it was used for water decontamination. The affinity of the cations to the PB absorbent was dependent on the hydration radius. The smaller the cation's hydration radius was, the more affinity to PB it had. Cesium's hydration radius (3.25 Å) was less than that of potassium (3.3 Å), sodium (3.6 Å), calcium (4.1 Å), and magnesium (4.25 Å). During multiple studies, the PB nanoparticles were altered using different methods. For example, Hara *et al.* made a thin layer of film by alkyl ligand-covered PB nanoparticles in organic solutions.[92] Glass and silica-based matrix nanocomposites along with PB nanoparticles have a 10 times greater rate of adsorption and three times higher capacity for adsorption than large PB particles. Torad *et al.* suggested that PB nanoparticles with a level of 330 m^2/g had more Cs ion-removal efficiency than commercial PB.[38] Omura *et al.* covered the nanoparticles of PB with ferricyanide anions. The small size of the PB nanoparticles caused an increase in the

surface-to-volume ratio followed by an increase in Cs uptake.[93] Sasaki *et al.* used PB altered by magnetite (PB-Fe_3O_4) to remove Cs from the water. The maximum level of Cs adsorption was 16.2 mg/g.[94] Ishizaki *et al.* created PB nanoparticles ($Fe_4(Fe(CN)_6)_3$) with several hydrophilic sites. Cs was adsorbed in great amounts by the proton removal reaction from this site.[18] The stoichiometric ratio of Fe:Cs in the PB particles and PB nanoparticles was, respectively, 8:1 and 1:1; therefore, the use of PB nanoparticles increased the Cs removal efficiency by a great number.[11] Thammawong *et al.* used magnetic nanoparticles (MNP) connected to PB to remove Cs.[17]

Magnetic particles are used to absorb Cs from a body. This compound has a high absorption capacity (96 mg Cs/g sorbent) and high distribution coefficient (3.2×10^4 ml/g) at 0.5 ppm Cs.[17] Despite the high efficiency of MNP in the removal of Cs, retrieving it from tap water, unlike magnetic particles, is not easy. Namiki *et al.* have used Prussian blueberry (PB-coated magnetic nanoparticles) to remove Cs from seawater.[1] The results show that Prussian blueberry (5 mg) decreases the Cs concentration from 150 ppm to 5 ppm. Also, on a large-scale, use of Prussian blueberry for 5 minutes reduces Cs concentration to 35%–35.6%.[1] Jang *et al.* have investigated the removal of Cs from aqueous solutions by PB graphene oxide hydrogel encapsulated within a PVA-alginate, in a fixed-bed column. PB nanoparticles were attached to the graphene oxide layers. The effect of pH on Cs removal (1 mmol concentration, flow of 0.83 ml/min, and 5 cm height) increased from 1 to 7, and the absorber reached a saturation point in less time. As PB dissolves in hydrogel in acidic environments, Cs absorption increases.[13] However, in an alkaline environment, Cs adsorption is reduced due to the formation of strong links between ferric ions and hydroxyl ions, which make band Fe-CN-Fe in PB (pH 7–9).[95] The initial increase of Cs concentration from 1 to 5 mmol and an increase in bed height from 5 to 20 cm increased the adsorption from 141.6 to 163.1 mg/g. Also, with the increase of the sorbent's particle size from 2 to 5 mm, the absorption capacity decreased from 161.6 to 130.96 mg/g. In a neutral pH flow rate, the bed height increased by reducing the initial concentration of Cs as well as the absorption efficiency.[13] The presence of PB components in tap water, such as binding polyvinyl alcohol and alginate, can endanger human health.[96] Chen *et al.* investigated the removal of the remaining Cs components and PB from the aqueous solution by PB and ion exchange. Two scenarios were performed. In the first one, the effect of ion exchange granules on PB was studied, and in the second, the ion exchange column, along with PB, in a large-scale water treatment plant were studied. The results showed that both methods could completely remove Cs (98% efficiency in a contact time of 50 seconds, and in 150-second contact time, there was lower than 0.01 μg/l limit of detection) and the remaining components of PB, such as ferricyanide ions and total cyanide.[97] The high solubility of Cs and its low surface charge had decreased its removal by the adsorbents via hydrophobic interaction or electrostatic interaction mechanisms.[98]

Conclusion

This study aimed to review the adsorption process for removal of Cs from water and aqueous solutions. The results showed that conventional water treatment methods could not efficiently remove Cs. The highest MAC of Cs was related to natural chabazite (273.24 mg Cs/g adsorbent), hollow PB nanoparticles (262 mg Cs/g adsorbent), PB-implemented non-woven fabric (260 mg Cs/g adsorbent), natural mordenite (254.7 mg Cs/g adsorbent), and PB granules (241mg Cs/g adsorbent). The minimum MAC of Cs was related to crushed granite (1.32 mg Cs/g adsorbent) and walnut shell (0.1mg Cs/g adsorbent). Several studies showed a high affinity of PB for Cs adsorption from aqueous solutions. On account of its high absorption capacity, natural chabazite should be paid more attention in the future research. It must be noted that the presence of absorbent residues in water is the most important health problem and must be taken into consideration in future studies.

Conflict of interest

The authors have no conflict of

interest to declare.

Author contribution

Y.FA conceived the study; Y.FA, M.SA, Z.BA, and N.AM designed the study protocol; H. KE, B. MO, and R. PO searched the literature; H. KE extracted the data; M.FE and G.OC analyzed and interpreted the data; Y.FA and M.AB wrote the manuscript; N. Am and B.MO critically revised the manuscript.

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