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Title page

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Adsorption of cesium on different types of activated carbon

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Abstract

The optimal conditions to remove radiocesium from water by adsorption on activated carbon (AC) were investigated. Two commercial ACs were compared to ACs prepared by steam activation of brewers' spent grain. The influence of pH and loading AC with Prussian Blue were studied. ^{134}Cs, measured by gamma-ray spectroscopy, served as a tracer for the Cs concentration. Column experiments showed that a neutral to acidic pH enhanced adsorption compared to high pH. Norit GAC 1240 had the highest adsorption capacity, 8.5 µg Cs g^{-1} AC for a column filtration. Sequential columns of Norit GAC 1240 removed 28.1 ± 2.8 % of Cs per column.

Keywords

Activated carbon
Adsorption
Radiocesium
Radioactive waste water

Environmental remediation
Low-level (radioactive) waste

1. Introduction

Produced only by anthropogenic sources, radioactive Cs isotopes are released into the biosphere by weapons testing, nuclear reactor accidents and controlled release into waste water streams. These releases combined with its relatively long half-life ($T_{1/2} \approx 30$ years), makes ^{137}Cs the major contributor to the long term environmental radiation dose received by humans and other organisms [1]. During the nuclear accident at the Fukushima nuclear power plant, the estimated release of ^{137}Cs into the environment amounted to more than 12 10^{15} Bq. Additionally, the short-lived ^{134}Cs (>12 10^{15} Bq, $T_{1/2} \approx 2$ years) and ^{136}Cs (>2 10^{15} Bq, $T_{1/2} \approx 13$ days) were also released, raising the level of activity in the drinking water above the legally permitted levels in the nearby areas [2,3]. One estimates that these atmospheric releases are only a fraction of the releases from the Chernobyl nuclear power plant during the accident in 1986 [2]. The activity of Cs radioisotopes poses a radiotoxicity risk caused by external radiation exposure and internal radiation damage after inhalation of contaminated air or intake of contaminated food or water [4,5]. However, stable Cs in these concentrations is not known to be harmful. Cs is very mobile in aqueous environments because of its high solubility, but it strongly binds to soils and minerals. This promotes accumulation of Cs radioisotopes and contamination of the food chain [5,4,6]. Treatment of radioactive waste water contaminated with Cs isotopes is a challenging research area in environmental radiation protection [7].

Recently, a wide range of low-cost adsorbents were investigated for the removal of Cs from wastewater because other methods such as chelation and precipitation are rather ineffective for the removal of trace amounts of Cs [8-10]. Studies have been carried out using different organic and inorganic ion exchangers, such as Prussian blue (PB) and other hexacyanoferrates (HCF) [11-13]. HCFs have a major disadvantage: they exist mainly as micro-particles (particle diameter <100 nm). These micro-particles are hard to filtrate from aqueous solutions and can clog a fixed bed reactor causing a significant pressure loss. Recent research has shown that adsorption of cesium is a promising remediation method for contaminated liquid wastes, if the operating costs can be kept sufficiently low [7,14,15]. Combining HCF (nano)particles with a carrier material having beneficial characteristics could provide a practical and efficient material for Cs removal [9,12,16,17]. Incorporation of HCF on biosorbents provides a solution for the filtration problems associated with unbound HCF [9,17]. The incorporation of these HCF in a porous material would lead to an increased amount of HCF for a specific volume, thereby increasing adsorption efficiency.

Activated carbon (AC), having high surface area and developed porosity is a low-cost and effective adsorbent for a wide range of pollutants [18,19]. AC has been used in research to remove radionuclides from waste water solutions in a relatively straightforward way compared to other methods [20]. AC has multiple surface functionalities, a high mechanical strength and a good resistance towards chemicals, heat and radiation [19,21]. Most of the AC produced presently is made by steam activation of mined coal. However, it might be economically interesting to produce AC from biomass by pyrolysis, followed by chemical or physical activation. During physical activation, a carbonised biomass is treated with a mildly oxidising gas at 750 – 900 °C to increase its porosity and surface area [18,22-25]. Carbonisation and activation of biomass has previously proven to produce economically valuable AC, if the source material has a consistent and lignin-rich composition [19,26-28]. Brewers spent grain (BSG) is an interesting source material for the preparation of AC because of its availability and high nitrogen content. This creates an in-situ nitrogenised AC, characterised by an increased amount of pyrrolic and pyridinic surface groups [29,30,27,31]. Because of changed acidic/basic surface characteristics and a more pronounced chemisorption mechanism, nitrogenised ACs have shown an improved adsorption towards multiple pollutants compared to normal AC [32-37]. Adsorption of Cs on ACs has been researched in the past, but with relatively high concentrations of Cs, generally 10 mg/L and higher [7,38,39].

In order to find an AC suitable for Cs removal from waste water, adsorption capacities for lower Cs concentrations are measured and presented in this paper. Furthermore, a simple HCF incorporation on the surface of the AC is tested, in order to find a synergistic effect between adsorption and the HCF ion exchangers. Focussing on low concentrations of Cs and different adsorption techniques, adsorption capacities are compared for different types of AC. A series of experiments was set up for the study in order to investigate the key adsorption parameters. This could provide good tools for future low-level adsorption experiments.

2. Experimental

The goal of this experiment was to study and optimise the removal of Cs from water using different ACs. To study the behaviour of the Cs, ^{134}Cs was used as a tracer. A standard Cs solution was irradiated to activate a fraction of the stable Cs to ^{134}Cs. The ^{134}Cs solution was brought in contact with AC under different experimental conditions. In this experiment the optimal settings for adsorption were tested by varying the AC, the pH and the adsorption technique.

Three adsorption techniques were evaluated: batch adsorption tests and two types of column tests. Batch adsorption tests were based on the equilibrium between the AC adsorbents (5 different types plus their modified form) and the Cs solution after 48 hours of shaking. Column adsorption tests resembled industrial adsorption filter systems and can be performed in two ways. Firstly, a single column containing the AC can be used multiple times to extract Cs from the same solution, until saturation of the AC surface is reached. This is referred to later on as a ‘single column’ experiment and is conducted with the 5 ACs. Secondly, a solution can be filtered through different sequential columns to remove Cs. This is referred to as ‘sequential column’ experiment and was performed only for Norit GAC 1240.

To prepare the solutions needed in the experiments, Milli-Q/Nanopure water and analytical grade reagents were used. All labware was cleaned with detergent and water prior to use. Glassware, centrifuge tubes and filtration columns were filled with a solution containing 20 mg L^{-1} of stable Cs having the same pH as the solution used in the experiments and left overnight in order to saturate their surfaces with stable Cs.

2.1 Activated carbon

Five different types of AC were used in the experiments. Three of them were prepared from BSG using a custom made pyrolysis/activation reactor as described previously in [27]. BSG was dried at 105 ± 5 °C for 24 h and sieved to obtain a particle size smaller than 2 mm. These three ACs were prepared at different temperatures, using different steam activation durations and quantities of Milli-Q water: ACBSG05 (800 °C/30 min/10 mL), ACBSG06 (850 °C/45 min/15 mL) and ACBSG07 (800 °C/45 min/15 mL). For comparison, the two most widely used commercially available ACs were used: Norit GAC 1240 (Cabot Corporation, Massachusetts, USA) and Filtrasorb400 (Chemviron Carbon, Seneffe, Belgium). Both of these ACs are made from steam-activated bituminous coal and have a more mesoporous structure. Their properties make them ideal for removal of a range of pollutants from waste water [40,41].

An important characteristic of AC is the point of zero charge (pH_{PZC}) as it determines at which pH the total surface charge of the AC is zero. A pH above the pH_{PZC} will cause the surface of the AC to be negatively charged, attracting the Cs cations to its surface. Solutions having $pH > pH_{PZC}$ will be tested to evaluate the influence of this attraction on adsorption.

The characteristics of the porosity of the AC were determined by analysis of the BET (Brunauer-Emmet-Teller) surface areas (total surface area S_{BET}, microporous surface area S_{micro} and exterior surface area S_{ext}), total porosity volume (V_T), micropore volume (V_{Micro}) and mesopore volume (V_{Meso}) by nitrogen (77 K) adsorption using an Autosorb AS-1 (Quantachrome, Düsseldorf, Germany) [42,43]. The micropores are characterised by the Dubinin – Radushkevich method (volume micropores V_{Micro}) and the t-plot method using the De Boer method (micropore surface S_{micro} and extremal surface S_{ext}). Before analysis the samples were outgassed for 16 h at 200 °C in high vacuum. Experiments were performed by 'Department of Chemistry', Laboratory for Adsorption and Catalysis, University Antwerp, Belgium.

Table 1 BET surfaces and pore volumes of the used ACs determined by nitrogen adsorption at 77 K

	S_{BET} **(m^2/g)**	S_{micro} **(m^2/g)**	S_{ext} **(m^2/g)**	V_T **(cm^3/g)**	V_{Micro} **(cm^3/g)**	V_{Meso} **(cm^3/g)**	L_0 **(nm)**	E_0 **(kJ/mol)**
ACBSG05	**686**	**422**	**100**	**0.335**	**0.274**	**0.060**	**0.9**	**23.5**
ACBSG06	**836**	**506**	**151**	**0.439**	**0.342**	**0.097**	**1.3**	**19.9**
ACBSG07	**758**	**461**	**121**	**0.382**	**0.304**	**0.078**	**1.0**	**22.2**
Norit GAC 1240	**1468**	**532**	**725**	**0.827**	**0.599**	**0.228**	**2.6***	**15.5**

Filtrasorb F400	**1483**	**833**	**432**	**0.859**	**0.604**	**0.256**	**2.6***	**15.5**

*** The empirical correlation (Stoeckli formula) is only valid for L_0 values between 0.5 and 2.0 nm**

For the ACs from BSG, the BET surface increases with increasing activation (from ACBSG05 to ACBSG07 and to ACBSG06). The obtained BET surface for the ACs from BSG is around half (686 – 836 m^2/g) of the BET surface from the commercial ACs (1468 -1483 m^2/g). Higher and longer activation times increase the BET surface, micropore (V_{Micro}), mesopore (V_{Meso}) and the overall pore volumes (V_T) of all the samples. The V_{Micro}/V_T ratio decreased from 0.81 to 0.78, indicating that the mesopores gain a larger contribution to the total pore volume by increasing activation. This is also observed in the broadening of the average micropore diameter (L_0). In contrast to the commercial ACs, the ACBSGs have average pore diameter that is 2 times smaller than the commercial ACs.

A rough estimation of the pore size distribution of the micropores (**Fig. 1**) is determined by means of the Density Functional Theory (DFT). The pore size distribution reveals that the ACBSGs consist of primarily pores with a diameter between 1 and 1.1 nm, followed by two secondary micropores 0.82 – 0.97 nm and 1.12 – 1.32 nm. In addition wider micropores are found of about 2 nm. The pore size distribution in the mesopore range shows four peaks: at 3.7 nm, 4.9 nm, 5.3 nm and 5.8 nm respectively. An increase in temperature and activation increases the amount of micropores and mesopores, without increasing their size. When comparing with the pore size distribution of the commercial ACs, a very discrete distribution is obtained by the activation and pyrolysis of BSG.

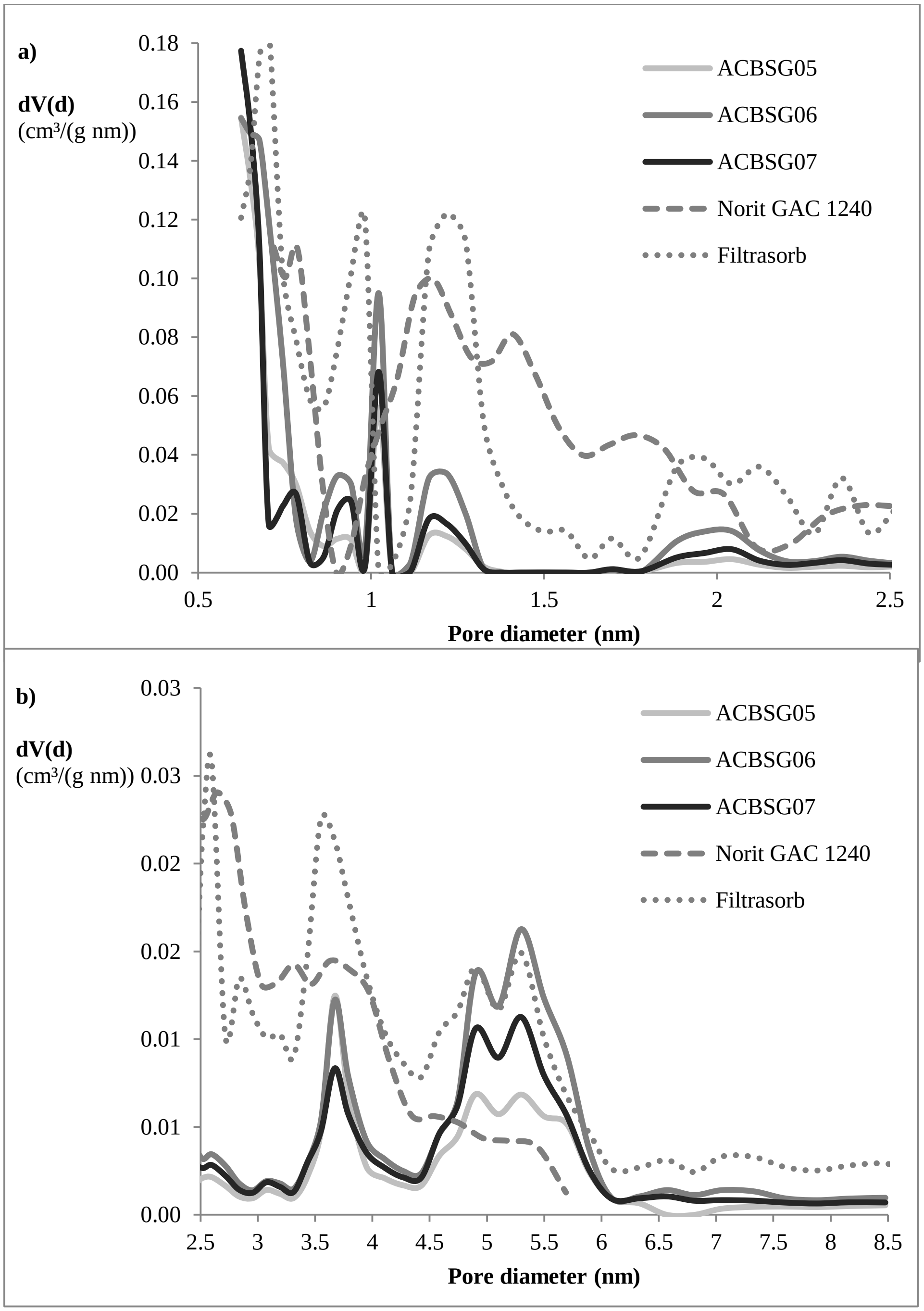

Fig. 1 Pore size distribution of the ACs determined by DFT method for a) micropores; b) mesopores (2.5 – 8.5 nm)

Prussian Blue (PB) was fixed on the surface of each of these five ACs to create an AC-PB combination to enhance the adsorption capacity. Therefore, 1 g of AC was mixed with 150 mL of saturated PB solution and shaken for 48 hours. The pH of the solution was set at 7, to prevent dissociation of the PB in an acidic or basic environment [44,45]. The AC was then filtered off using ashless Whatman filters, washed with water and dried at 105 ± 5 °C for 24 h. The PB concentration of the solution before and after adsorption was obtained indirectly by measuring the iron concentration in the solutions using ICP-AES (Optima 3300 DV, Perkin Elmer, Massachusetts, USA). The mass difference of PB was supposed to be bound to the AC. The mass of PB adsorbed on the surface of the AC is an important parameter to determine the extra amount of Cs that can be adsorbed. Both 5 ACs and 5 AC-PBs were used in the batch adsorption experiments.

Table 2 displays the mass of PB adsorbed on the AC expressed in mg g^{-1}. The observed differences in adsorbed amount might be related to the pore size distribution, allowing PB to move more freely in Norit GAC 1240, Filtrasorb 400 and ACBSG06. The latter was activated in the most severe conditions (850 °C/45 min/15 mL), which led to an increase in overall pore sizes, as described above, increasing the mobility of the colloidal PB molecules.[46]

Table 2 Mass of PB adsorbed on the five different activated carbons used in this study

Type of AC	mg PB g^{-1} AC
Norit GAC 1240-PB	13.9 ± 0.1
Filtrasorb 400-PB	10.1 ± 0.1
ACBSG05-PB	1.6 ± 0.1
ACBSG06-PB	15.3 ± 0.1
ACBSG07-PB	3.3 ± 0.1

2.2 Solutions

In order to be able to monitor the behaviour of Cs in these experiments, a 1000 mg L^{-1} $CsNO_3$ standard solution was irradiated for 21 hours in the neutron flux of BR-1 at SCK·CEN ($\varphi=3\ 10^{11}$ n cm^{-2} s^{-1}, $\sigma=30\ 10^{-24}$ cm^{-2}) to activate part of the Cs to ^{134}Cs. The ^{134}Cs in the irradiated solution served as a tracer for the total Cs. The undiluted solution had an activity concentration of 50.93 ± 0.74 10^{3} Bq g^{-1}. The radiopurity of it was checked by measuring it on a HPGe detector. No impurities could be identified.

The solution was diluted to approximately 1:1000 in three steps. Dilution factors were determined gravimetrically and checked by measuring the activity of ^{134}Cs in the solution using an ionisation chamber and a well-type NaI(Tl) detector.

Previous work showed that the pH_{PZC} of AC from BSG ranges from 10.6 to 10.8, and those of the used commercially available ACs from 11.5–11.7 [27]. Therefore the pH of the working solutions was adjusted to 7, 10 and 12 using ammonia. These working solutions had a ^{134}Cs activity concentration of 59.24 ± 0.70, 59.67 ± 0.70, 60.44 ± 0.71 Bq g^{-1} corresponding to a Cs concentration of 1.16, 1.17 and 1.19 mg L^{-1} at pH 7, 10 and 12, respectively.

2.3 Gamma-ray spectrometry

The activity of the samples was determined by 4π gamma counting using a 20 x 20 cm NaI(Tl) well-type detector with a well-diameter of 25.4 mm and well-depth of 134.0 mm. In the NaI(Tl) well detector all the signals above the lower energy threshold of 50 keV were counted. All measurement results were corrected for background and decay. The total efficiency of the well-type NaI(Tl) detector was calculated using Monte Carlo simulations with the EGSnrc-code. The calculations were done using the same lower threshold of 50 keV. Furthermore, the different filling heights of solution in the used centrifuge tubes were individually modelled. The calculated efficiencies were close to 100 % and showed a variation smaller than 1 % between empty and completely filled tubes.

A gamma-ray spectrum of the ^{134}Cs solution obtained with the well-type NaI(Tl) detector is displayed in **Fig. 2**. The lower energy threshold of 50 keV is clearly visible. The spectrum shows the transitions at 605 keV (p=98.21, $\gamma_{1,0}$) and 796 keV (p=85.73, $\gamma_{3,1}$) [47]. The peak at 1401 keV is the sum peak of these transitions, caused by the simultaneous detection of both gamma rays. At 1970 keV the gamma rays of 569, 605 and 796 are collected simultaneously.

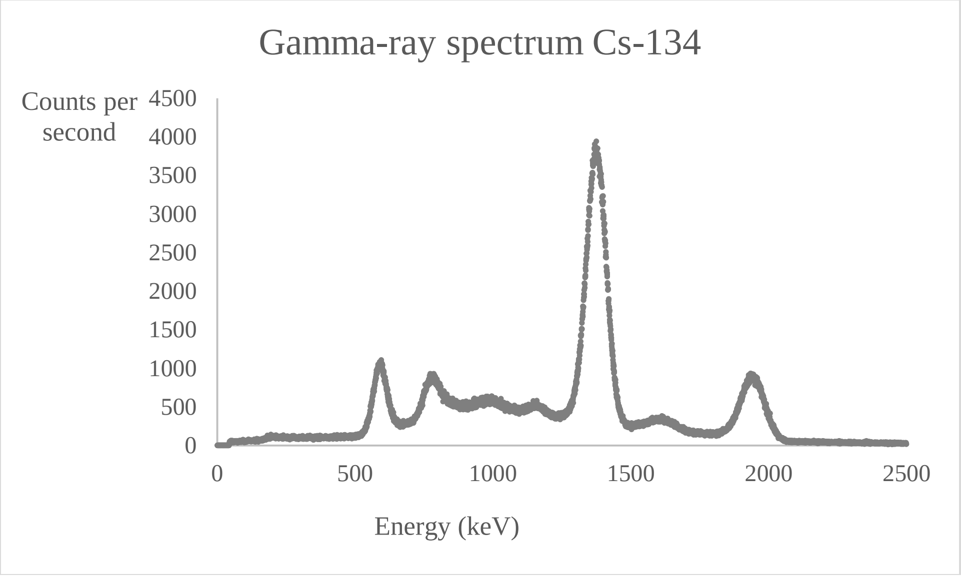

Fig. 2 Gamma-ray spectrum of a ^{134}Cs solution measured in the well-type Na(Tl) detector

2.4 Batch adsorption experiment

The batch adsorption experiments were conducted using all 10 types ACs, i.e. 5 ACs both with and without fixed PB. Each of these ten ACs was tested for adsorption using three ^{134}Cs solutions of different initial pH (7-10-12) and similar activity.

Between 20 and 30 mg of AC and approximately 9 mL of Cs solution (approximately 60 Bq g^{-1} ^{134}Cs, corresponding to 1.2 mg L^{-1} Cs) at the desired initial pH (7, 10 or 12) were gravimetrically added into different centrifuge tubes (VWR High Performance 15 ml, VWR International, Leuven, Belgium). After shaking the tubes for 48 h in a Maxi-Mix III Vortex Mixer (Thermo Scientific, USA), each solution was filtered through a 75 mm funnel (VITLAB, Großostheim, Germany) with an ashless Whatman filter and the filtrated solution was stored in a second centrifuge tube. The filter containing the AC was dried and stored in a third centrifuge tube. All three centrifuge tubes (the empty centrifuge tube, the filtered solution and filter paper with AC) were measured in the well-type NaI(Tl) detector. This test was conducted in triplo for each type of AC. Adsorption capacities at equilibrium (q_e, in mg/g) were calculated as follows:

$$q_\mathrm{e} = A_\mathrm{filter}\, k_\mathrm{ac} / m_\mathrm{AC}. \quad (1)$$

Were A_filter is the measured activity in the filter (in Bq), k_ac is the recalculation constant for activity to mass (in mg/Bq) and m_AC is the mass of the AC used (in g).

To prove there were no losses of ^{134}Cs during the experiment, a recovery experiment was set up. This experiment was conducted in exactly the same way prior to the batch adsorption experiment. Recovery rates were calculated for three ACs as the ratio between the activity of the solution in the centrifuge tube prior to shaking and the sum of the activities measured in the three centrifuge tubes after the adsorption experiment. A maximum of 0.7 % deviation from 100 % was found, indicating that the losses of activity throughout the adsorption procedure were minor.

2.5 Single column experiment

Adsorption of Cs on AC can be applied to both surface water (neutral to slightly acidic environment) and liquid waste treatment in decontamination units, where pH conditions can be more extreme. In order to test samples resembling the conditions described above and to find an optimal pH to promote column adsorption, solutions of different pH were tested: one acidic, one neutral and 2 basic. As a reference AC, Norit GAC 1240 was chosen for this experiment. Bio-rad Poly-Prep Chromatography Columns (0.8 x 4 cm) (Bio-Rad, California, USA) were filled with approximately 0.7 g of Norit GAC 1240 and pre-wetted with water. 11 mL of approximately 37 Bq g^{-1} ^{134}Cs solution at pH 4, 7, 10 and 12 was poured over these columns and collected, by gravity, in a centrifuge tube. The activity of the collected solution, as well as the activity remaining in the empty centrifuge tube, were both measured in the well-type detector. The collected solution was then poured over the column again. This cycle was repeated five times. Each test was conducted in duplo. After five cycles the column was measured in the well-type detector after air drying for 48 hours. Adsorption capacities (q) were calculated as mg Cs (calculated from the column activity) per gram of AC.

On the basis of the first tests the most effective pH was determined, 5 different ACs (Norit GAC 1240, Filtrasorb 400, ACBSG05, ACBSG06, ACBSG07) were used in order to compare their performance. The column adsorption procedure was identical to the one described above. It was performed using 11 mL of approximately 37 Bq g^{-1} ^{134}Cs solution at pH 7.

For the column adsorption experiment a recovery experiment was also conducted. For this purpose, two columns filled with approximately 0.7 g of AC (Norit GAC 1240 and ACBSG07) were used. 11 mL of containing approximately 37 Bq g^{-1} of ^{134}Cs was then poured over each column and collected in a centrifuge tube. This process was repeated 5 times. The centrifuge tubes and column were measured as described above. The average recovery rate showed no loss during the column test.

2.6 Sequential column experiment

In this part of the experiment five columns were filled with approximately 0.7 g of Norit GAC 1240. 12 mL of approximately 37 Bq g^{-1} Cs solution at pH 7 was poured over the column and collected. After measuring the collected solution, it was poured successively over the four remaining identical columns and the activity of the solution was measured after each filtration. This test was conducted in duplo.

3. Results and discussion

3.1 Batch adsorption experiment

Batch adsorption tests using different ACs could reveal the difference between the commercial ACs and the AC from BSG. The influence of PB adsorption prior to Cs adsorption and the influence of pH on the amount of adsorbed Cs were also determined.

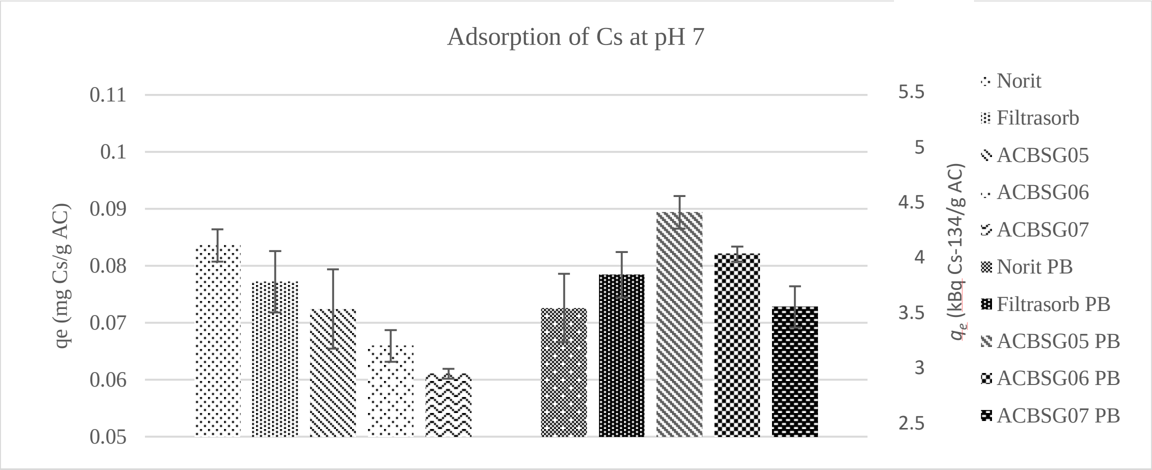

Fig. 3 Adsorption capacities (q_e) of Cs on different ACs during batch adsorption at pH 7

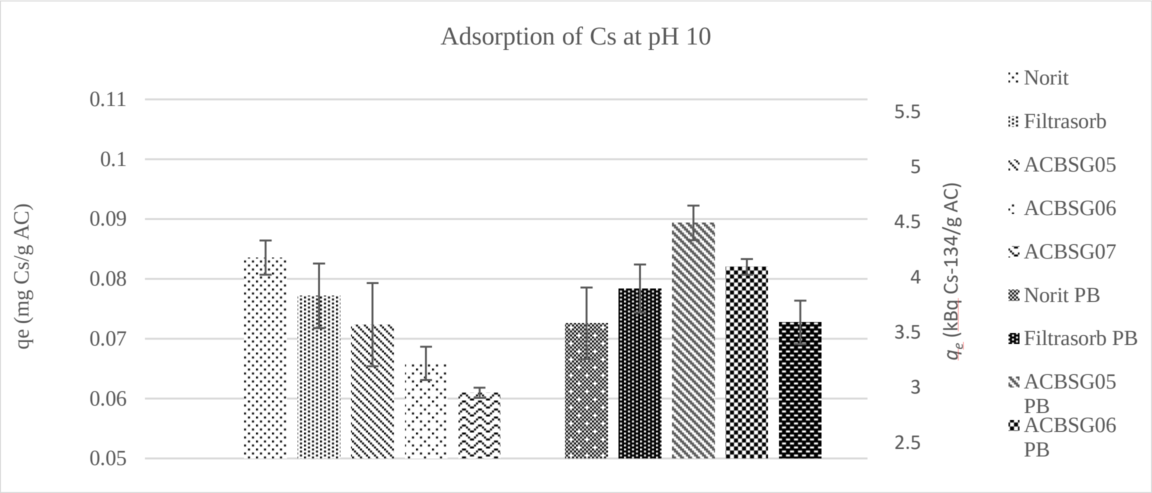

Fig. 4 Adsorption capacities (q_e) of Cs on different ACs during batch adsorption at pH 10

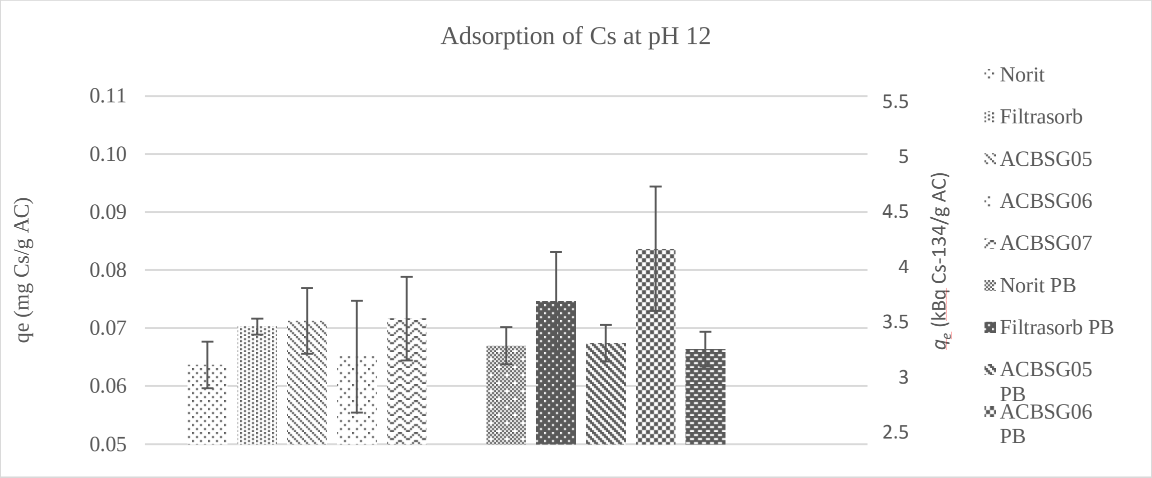

Fig. 5 Adsorption capacities (q_e) of Cs on different ACs during batch adsorption at pH 12

The results of the batch adsorption experiments are displayed in **Fig. 3-5.** The untreated ACs are plotted on the left and the ACs with PB adsorbed on the right side. Error bars show the combined standard uncertainty. Statistical measurement uncertainties were calculated and proven to be insignificant compared to the standard deviations between the 3 different repeats of the experiments. For each pH the average activity concentrations and Cs concentrations before and after adsorption are displayed in **Table 3**. The average removal percentage is displayed at the different pH settings. Adsorption capacity at equilibrium is expressed as q_e (mg Cs adsorbed per gram AC).

pH	Activity concentration before	Cs concentration before	Activity concentration	Cs concentration after	Removal percentage

	adsorption (Bq g^{-1})	adsorption (mg L^{-1})	after adsorption (Bq g^{-1})	adsorption (mg L^{-1})	
7	59.24 ± 0.70	1.16 ± 0.01	48.63 ± 1.86	0.95 ± 0.04	18.3 ± 3.0
10	59.67 ±0.70	1.17 ± 0.01	47.11 ± 1.25	0.93 ± 0.02	21.0 ± 1.7
12	60.44 ± 0.71	1.19 ± 0.01	49.24 ± 1.18	0.96 ± 0.02	18.5 ± 1.2

Table 3 Average measured (activity) concentrations before and after batch adsorption with calculated average removal for each tested pH

The best adsorption capacity (q_e) was obtained at pH 10, although the differences between adsorption of solutions of different pH were not significant. At pH 12, all ACs were negatively charged, but this did not increase the adsorption of Cs on the AC. This may have been caused by the competition of adsorption between Cs and the high amount of ammonia present in the solution. The binding of PB on AC prior to Cs adsorption did not significantly promote the adsorption at any of the tested pH values. Furthermore the dissociation of unbound PB happens above pH 8. This can explain the fact that AC with PB shows the same q_e values at pH 10 and 12 as for the unloaded AC. This experiment shows that the ACs have similar adsorption capacities in different circumstances. A removal of about 20 % is limited compared to the results obtained by ion exchangers, where removal rate of above 70 % are easily reached [48,16]. Some manuscripts report extremely low [39,49] removal percentages for Cs using AC. Kimura et al. [15] reported removal percentages up to almost 100 %, but used AC dosages were approximately 30 times higher than in this manuscript. The ACs investigated seem to have an interesting affinity for Cs, even at lower dosage.

3.2 Single column adsorption

3.2.1 pH selection

Results of the column filtration of solution at different pH (4-7-10-12) are shown in **Fig. 6**.

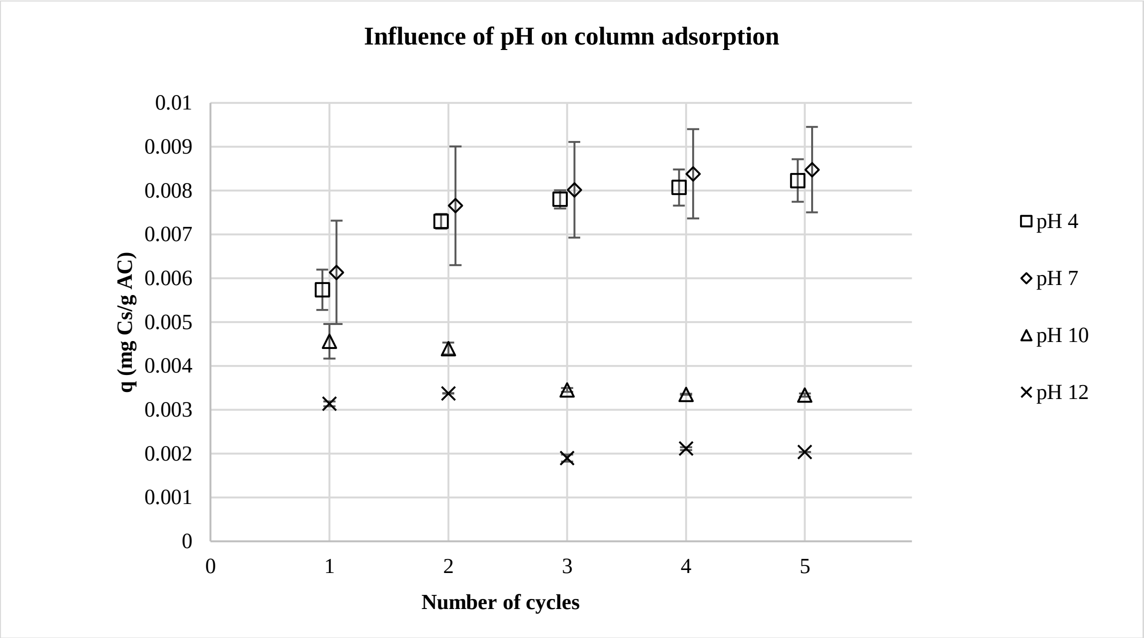

Fig. 6 Adsorption capacities after 5 cycles of adsorption on a column filled with Norit GAC 1240 using solutions with different pH. Data points have been artificially separated to enhance visibility.

For pH 4 and pH 7, the adsorption capacity (q) increased with each cycle with a maximum q_e of about 0.0083 mg g^{-1} Cs on Norit GAC 1240. A higher pH drastically lowered the maximum adsorbed amount of Cs on the AC, to a q_e of 0.0033 mg g^{-1} at pH 10 and 0.0020 mg g^{-1} at pH 12 after 5 cycles. Additionally q seems to decrease with each additional cycle. This effect was probably due to the competition of adsorption by the ammonia ion in the solutions, which explains why the lowest adsorbed amount is found at the highest pH. The competition between Cs and the ammonia ion may wash out the Cs already adsorbed on the AC, decreasing q after three cycles. A neutral to slightly acidic environment enhances Cs adsorption in comparison to a strongly basic environment.

3.2.2 AC selection

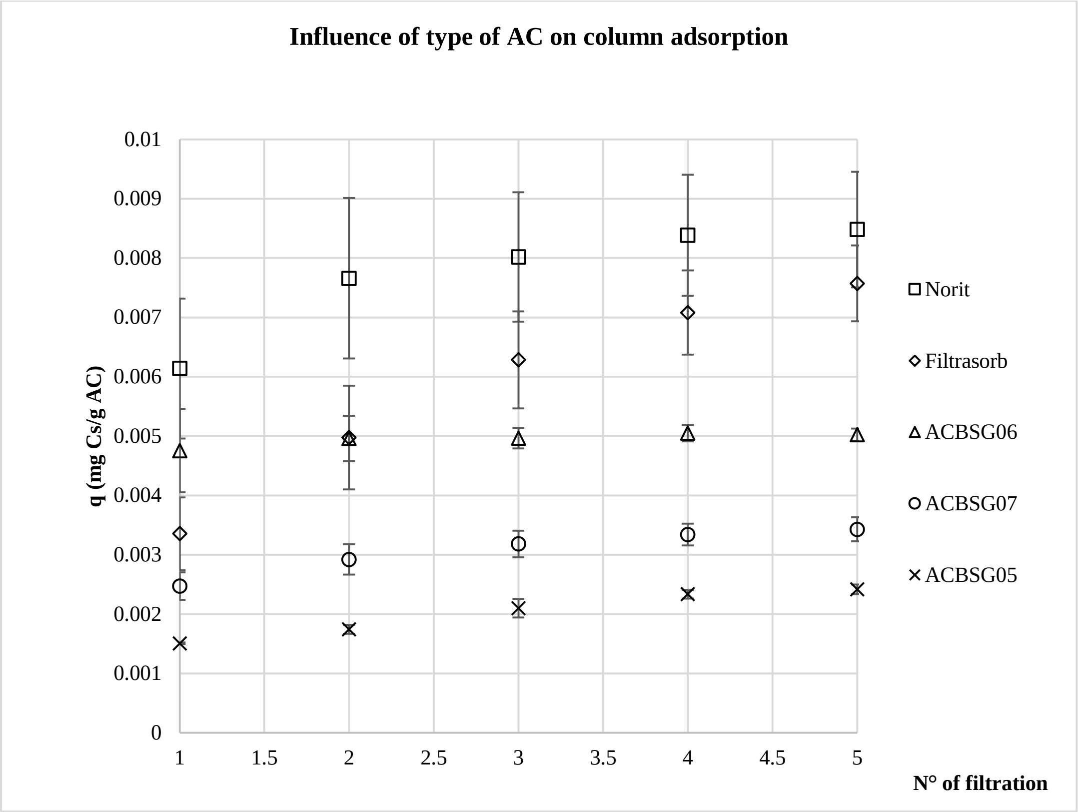

Fig. 7 Adsorption capacities after 5 cycles of adsorption on 5 ACs using a solution of pH 7

The commercial ACs show a significantly higher adsorption capacity *q* of Cs compared to the ACBSGs. Norit GAC 1240 reached its equilibrium adsorption capacity (q_e of 0.0085 mg g^{-1}) after 4 cycles, while *q* on Filtrasorb 400 still rose to 0.0075 mg g^{-1} after 5 cycles. Both commercial ACs are in granular form, making a better interaction between the surface of the AC and the Cs solution possible compared to the three ACBSGs, having a smaller particle size. The ACBSGs tended to stick together in the column with air still present in open spaces, causing possible channel formation. This resulted in a less efficient interaction between AC surface and solution. Additionally, this caused a limited contact time and smaller contact area for the ACBSGs. All 3 ACBSGs reached their equilibrium adsorption capacities after 2-3 cycles. ACBSG06 performed better compared to ACBSG05 and ACBSG07 (a *q* of 0.0050 mg g^{-1}, compared to 0.0024 mg g^{-1} and 0.0034 mg g^{-1} respectively), possibly due to the higher activation temperature used, creating a slightly wider pore distribution thus increasing the mobility of the Cs ions in aqueous solutions.

3.4 Sequential column adsorption

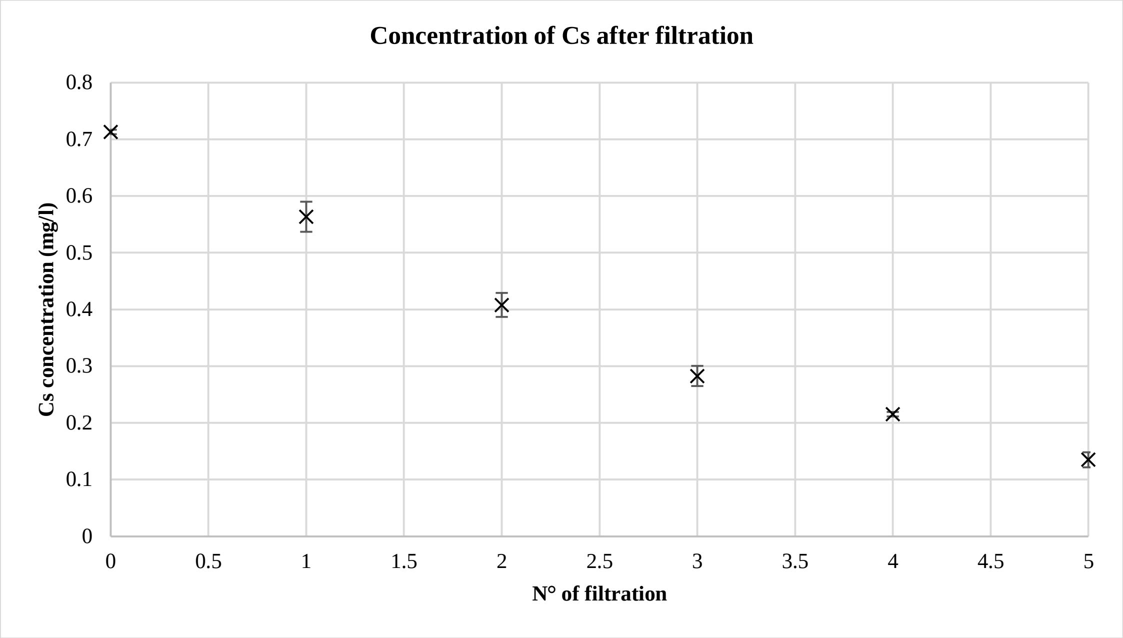

Fig. 8 Decrease in Cs concentration after sequential column adsorption with Norit GAC 1240 GAC1240

Passing the solution through five columns filled with Norit GAC 1240 decreased the concentration from 36.3 ± 0.3 Bq g^{-1} (0.713 mg L^{-1} Cs) to 6.8 ± 0.9 Bq g^{-1} (0.135 mg L^{-1} Cs). For this test, the decrease showed a removal rate of 28.1 ± 2.8 % of the initial Cs concentration per filtration. This result indicated that further removal of low levels of Cs is possible using sequential filtration steps. No equilibrium was reached during this experiment, as displayed in **Fig. 8**.

Conclusions

This work studied the removal of low concentrations of Cs from aqueous solutions by AC. For measurement ,^{134}Cs served as a tracer for the total Cs concentration. A standard solution of Cs was irradiated in a neutron flux to provide a radiopure ^{134}Cs solution of which both the exact concentration of Cs and activity of the ^{134}Cs tracer were known. Batch experiments using a variety of ACs adsorbing Cs from solutions with different pH showed no significant difference between the adsorption capacities at equilibrium for Cs (expressed as q_e values in mg Cs per gram AC) on either commercial AC or AC from BSG. Also the difference in adsorption between the Cs solutions of different pH was not significant. Binding PB on the ACs prior to the adsorption of Cs showed no significant effect on the q_e for any of ACs. Because batch experiments revealed no difference between the ACs, column experiments were conducted. A column experiment with Norit GAC 1240 using 4 solutions with a different pH showed that a neutral to slightly acidic pH increased the adsorption of Cs. At a higher pH the effect of the competition with ammonia ions caused a decrease of q_e. For the lab-scale ACs the ACBSG06 (the AC activated at the highest temperature) reached the highest q_e. Both commercially available ACs (Filtrasorb 400 and Norit GAC 1240) had even higher q_e values. This was caused by the difference in physical properties of the AC. Using sequential column adsorption to remove low concentrations of Cs from aqueous solutions led to promising results, as a steady removal rate per cycle step was observed. An equilibrium state is expected to be reached after several stages, where further removal is ineffective. A study will be performed to confirm this. The experiment showed that AC has potential as a low-cost and effective adsorbent for removal of low Cs concentrations. Removal percentages and adsorption capacities were comparable or higher than in relevant literature. Feasibility studies on the application of the method described in this paper in the nuclear industry are planned to be conducted.

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