

THESIS

STUDY OF HEXACYANOFERRATE(II) NANODIAMOND ADSORBENT FOR THE PRECONCENTRATION OF
CESIUM AND SIZE CATEGORIZATION OF RADIOCESIUM PARTICLES FROM CONTAMINATED SOIL

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ABSTRACT

STUDY OF HEXACYANOFERRATE(II) NANODIAMOND ADSORBENT FOR THE PRECONCENTRATION OF CESIUM AND SIZE CATEGORIZATION OF RADIOCESIUM PARTICLES FROM CONTAMINATED SOIL

High levels of radioactive cesium (RCs) were released into the environment as a result of the Fukushima Dai-ichi Nuclear Power Plant accident in March 2011. Wide areas, including residential and farming spaces, were contaminated to varying degrees with ^{134}Cs and ^{137}Cs which has presented complications with soil and water utilization for personal use. The current decontamination efforts used to remove RCs from the topsoil have been costly and created new challenges in storing low-level radioactive waste. Meanwhile, determining low levels of radioactivity in the environment is a challenge in itself, since changes in sample composition and self-shielding coupled with the sample amount required to meet low minimum detection limits lead to inaccurate measurements. This project investigates the use of a hexacyanoferrate(II) nanodiamond adsorbent to preconcentrate dissolved Cs in contaminated solutions in order to improve the counting efficiency and accuracy for low activity samples. Also, the determination of the size of particle attached radiocesium from previous soil samples that have undergone sequential extraction was attempted in order to study how particle size affects migration and deposition behavior. The majority of the preconcentration studies yielded over 98% ^{137}Cs adsorption. Further investigation into preconcentration studies is recommended to test adsorbent performance in more dynamic conditions.

Meanwhile, activity measurement of 0.45 μm , 0.2 μm , and 0.1 μm filters for Cs-137 indicated that the vast majority of the extracted RCs in the fractions obtained by sequential extraction was in a

dissolved form. Therefore, the RCs that was extracted from the soil is not present in the form of microparticles with an activity greater than 5 Bq.

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CHAPTER 1: INTRODUCTION

1.1 Background

This year marks the 10th anniversary of the Fukushima Nuclear Power Plant (FNPP) accident whose impacts are still affecting lives in Japan today. Wide areas, including residential and farming spaces, were contaminated to varying degrees with radioactive nuclides, the most prevalent being I-131, Cs-134, and Cs-137. Release calculations estimate about 1.5×10^{17} Bq of I-131 and 1.2×10^{16} Bq of Cs-137 were discharged into the atmosphere. (Chino et. al, 2012) Surface contamination calculations estimate that about 80% of the Cs-137 remains in the environment. (Tsukada, 2021)

Radioactive ^{137}Cs is formed by the fission of U-235, the isotope used to enrich the FNPP's uranium fuel which promotes sustainable nuclear reactions. Upon neutron capture, U-235 will split and causes a large release of energy, neutrons and two fission fragments. There is about a 6.2% chance (cumulative fission yield) that one of these two products is Cs-137, a radioactive isotope of cesium. This occurrence is relatively large compared to the formation of other fission product which occurs at fractions of a percentage. Other isotopes of Cs, such as Cs-135, are also produced by fission but at a much smaller percentage of $4.9 \times 10^{-4}\%$. (IAEA, n.d.) Cs-134 primarily is produced from the neutron capture of stable Cs-133. Cs-134 and Cs-135 play an important role in identifying the source of radiological release, as their ratio to Cs-137 will vary depending on fuel burnup. (Yang, Tazoe, and Yamada, 2016) In the environment, radioactive Cs presents a radiological hazard since all isotopes release energetic radiation during decay, have long half-lives, Cs-137 ($t_{1/2} = 31.7$ years), Cs-134 ($t_{1/2} = 2.06$ yr) and Cs-135 ($t_{1/2} = 2.3 \times 10^6$ yr) and are biological analogues to potassium.

Both particulate and dissolved Cs has been observed in the environment as a result of releases from the reactor accident. The majority of Cs will migrate to bodies of water by surface runoff, however,

both particulate and dissolved Cs can be adsorbed by soil and sediment layers, presenting a contamination hazard while causing local radiation levels to rise. Cs has also been observed to accumulate in rice paddy fields even when the Cs concentration in the irrigation source was below the concentration limits for drinking water. (Shin 2015) This presents another hazard as dissolved fractions can be up taken by plants and incorporated into the food chain. The remaining large amounts of RCs present near the powerplant has delayed resettlement and agricultural activities.

Current decontamination efforts used to remove RCs from the soil has been costly and created new challenges in storing and handling radioactive material. As of 2015, over 28 million cubic meters of topsoil has been striped. (Tanaka, 2015) The volumetric amount of removed soil has put pressure on the Japanese government to secure space for interim storage facilities. Also, the possibility of recontamination of agricultural areas from irrigation waters is another challenge faced in decontamination. (Shin, 2015) Investigation into more efficient Cs removal and understanding of Cs transport in the soil, could lead to more efficient decontamination efforts.

1.2 Cs Preconcentration

Every analyst faces certain challenges when attempting to measure RCs in the environment. Variations in sample collection and composition will affect counting geometry and efficiency and can therefore give rise to inconsistent results. Low levels of activity may go undetected due to self-shielding of the sample. Samples with small amounts of activity require preconcentration of the nuclide due to the large sample amount required in order to obtain confident measurements. A highly regarded standard for Cs preconcentration from aqueous samples is based on ammonium molybdophosphate (AMP) precipitation. (Daum, 2018) While AMP is an effective method, it is time intensive and difficult. Many other methods of concentrating RCs, primarily employed in pharmaceutical and environmental settings, are discussed next chapter. Of the methods, the ones utilizing Prussian Blue (PB) as a Cs adsorbent are of interest to this work due to their simplicity and effectiveness at collecting Cs from

aqueous samples. A newer nano scale form of the adsorbent, which utilizes the nanodiamonds to carry PB, has yielded exceptional sorption results, however previous studies were not carried out with radioactive cesium. The large surface area to volume ratio of the compound shows promise at concentrating Cs for low level activity measurements. This project aims at investigating the properties of a nanodiamond PB adsorbent under various experimental conditions.

1.2.1 Prussian Blue

Prussian blue is iron(III) hexacyanoferrate (II) with a formula of $\text{Fe}_4[\text{Fe}(\text{CN})_6]_3$, and it is primarily used as a blue pigment in dyes. The complex contains iron in both the (+II) and (+III) oxidation states. (ACS, n.d.) Potassium hexaferrocyanide, $\text{K}_4[\text{Fe}(\text{CN})_6]$, is a related compound to PB which has been shown to be effective at adsorbing Cs. (Nielsen, 1990) While the exact mechanisms for Cs adsorption onto insoluble forms of PB are unclear, a combination of ion exchange, physical adsorption, and mechanical trapping are believed to drive the action. (Estelrich and Busquets, 2021) The Cs would either exchange with a hydrogen or hydronium ion that are coordinated around the PB molecule or exchange with a monovalent cation, such as potassium, within the crystal lattice. Incorporation into the crystalline matrix is also possible due to electrostatic force. (Faustino, 2008)

Potassium ferrocyanide, which will be referred to as Prussian blue (PB) through the rest of this document, exists as a very fine powder and is soluble in water. In this form, Cs adsorption is driven by the ion exchange with the potassium ion. (Estelrich and Busquets, 2021) When used alone, the material will not completely precipitate from a solution after adsorbing Cs. (Matsumoto, 2018) Therefore, the PB must be either transformed or bonded to a host which will allow it to be more easily recovered from liquid samples.

1.2.2 Nanodiamonds

Nanodiamonds are nano-scale diamond particles that were originally produced during an explosion containing carbon. While many production methods are utilized today, the properties and availability of nanodiamonds provide a great option for research. Nanodiamonds have a high biocompatibility and are less toxic than other nanocarbons. (Schrand, 2009). This is a beneficial feature for environmental and biological applications. Due to their small size, nanodiamonds have high surface area to volume ratios. Their surfaces can also be conditioned to fulfill specific roles in experiments and applications. (Mochalin, 2021) In this study, nanodiamonds were used as the substrate to adhere the PB particles in order to extract cesium from water samples. The nanodiamond's surface was oxidized via heat treatment and then treated with copper chloride (CuCl_2) prior to addition of the PB.

1.3 Cs Particles

Unique particulate forms of Cs have been observed from the fallout which are spherical in shape and have a glassy composition. These particles are not easily dissolved in water and are believed to be retained longer in the environment. (Adachi, 2013) Since the health effects and behavior of these particles are not well known, they pose concern on the long-term impact on the environment. The second part of this work aims to further investigate the presence and properties of radiocesium bearing particles from previously collected and treated soil samples.

1.3.1 Soil Core Samples

Eight soil samples were collected from the difficult to return zone in the Fukushima Prefecture in 2013. The location of the sample sites is shown on Figure 1. Each core was 15 cm long and was collected using an AMS soil core sampler. The cores were cut into 2.5 cm segments and frozen until November of 2017. Then, the segments were dried at 106°C for 12 hours and massed. The segments were counted for radioactivity using a sodium iodide detector. The distribution of Cs-137 in the soil cores is shown in

Figure 2. The two most contaminated samples underwent a five-step sequential extraction, detailed below.

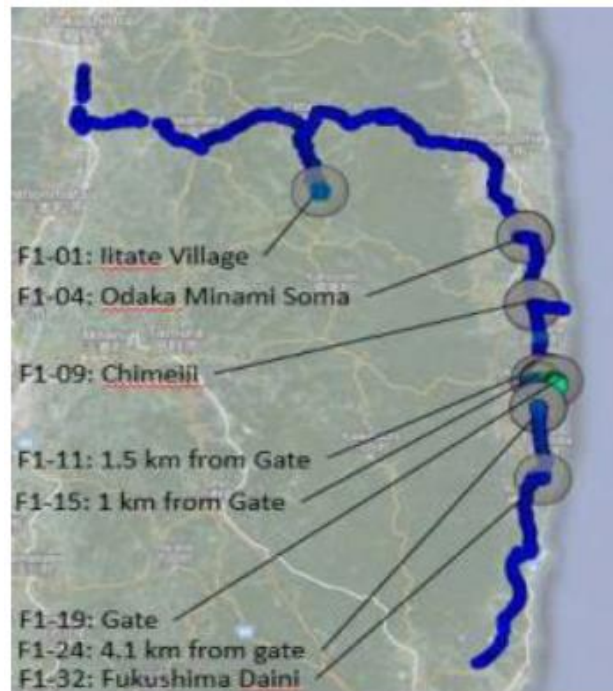


Figure 1. Sample locations of the soil cores taken in 2013. (McNabb, 2019)

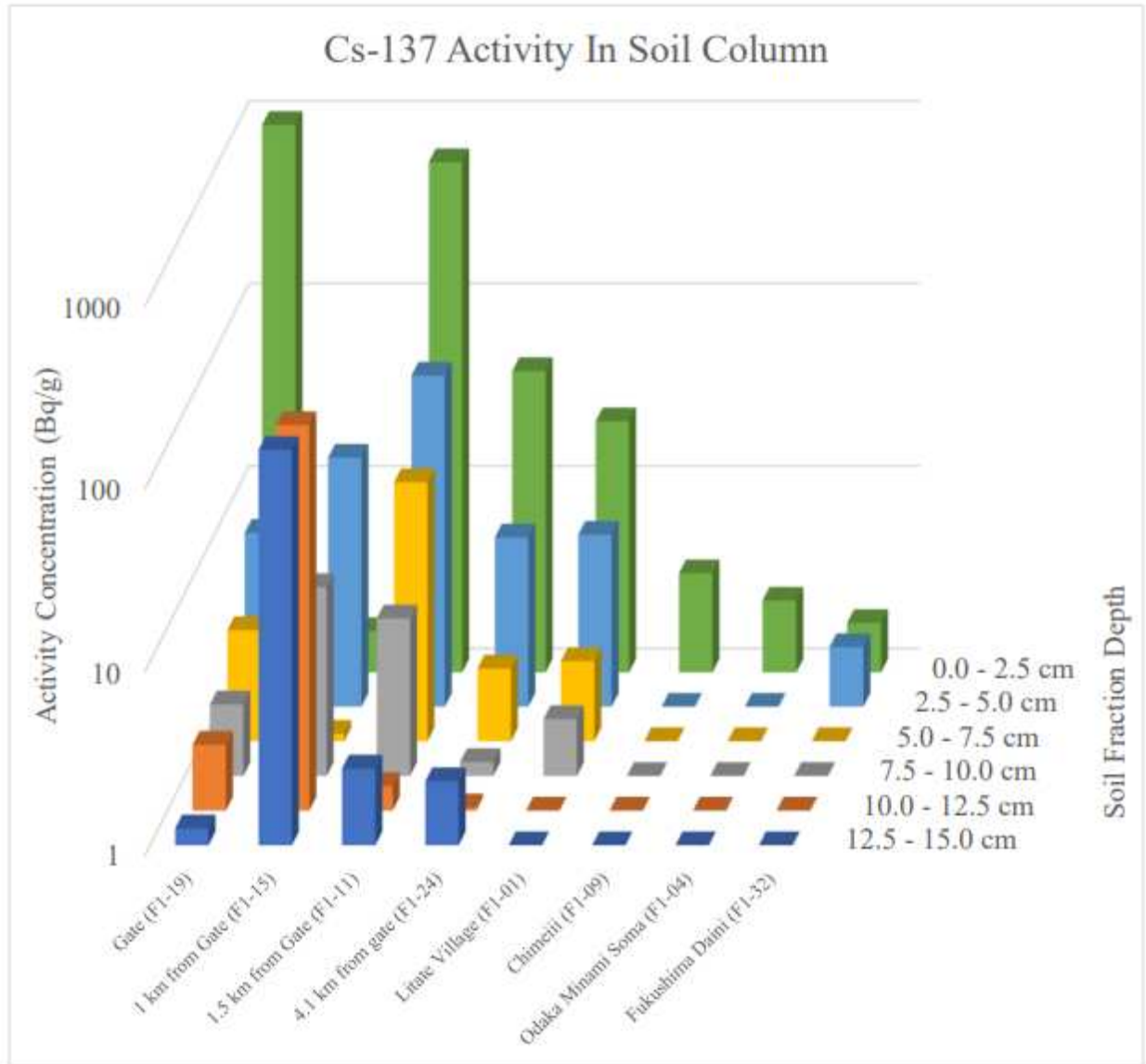


Figure 2. Cs-137 activity profile of the soil cores collected from the difficult to return zone in Fukushima Prefecture. (McNabb, 2019)

1.3.2 Sequential Extraction

The two most contaminated soil cores underwent a 5-step sequential extraction performed by McNabb (2019). Sequential extraction aims to separate an element of interest by dissolving specific host phases of the soil in a progressively aggressive succession. In this case, the process focused on the tightly bound Cs found in clay minerals in hopes of learning about its bioavailability and movement. The procedure targets the exchangeable fraction, carbonate bound, Fe/Mn oxide bound, organic bound, and

persistently bound fractions which could bind Cs. The reagents used, concentration, temperature, and duration held at that temperature are summarized in Table 1.

Table 1. Sequential Extraction Summary

Fraction	Reagent	Temperature (°C)	Duration (h)
1	0.1M MgCl ₂	25	1
2	1.0M NH ₄ Ac in 25% (v/v) HAc	50	2
3	0.1M NH ₂ OH•HCl in 25% (v/v) HAc	70	6
4	30% H ₂ O ₂ in 0.02M HNO ₃	70	3
5	4M HNO ₃	90	4

While the sequential extraction was successful at removing Cs from the soil, no distinction in Cs form was made in the extracted proportions. It is desirable to further investigate the composition and chemical form of Cs in the extracted fractions to either confirm or rule out the presence of particulate Cs.

2.1 Cesium and Clay Chemistry

The fate and transport of Cs in the environment is affected by numerous factors. Key components include soil mineralogy, presence of clay minerals, the weathering state of the minerals, presence of organic materials and soil chemistry. The physical and chemical forms of Cs also affect transport since particles are not as easily dissolved in water and will move differently than soluble forms of Cs. (Adachi et. al, 2021)

It is well known that clay minerals, particularly those with a 2:1 layer structure, have a strong affinity to bind Cs. Minerals belonging to this group include vermiculite and illite. Clay minerals can be further categorized as expanding or non-expanding based on the ability of their interlayer to expand in the presence of water. The expansion is primarily caused by the hydration of exchangeable cations that are in between the negatively charged layers. (Young, 2000) Expanding clay minerals have a higher cation exchange capacity and surface areas, which increases their affinity to adsorb Cs. (Park 2019) In non-expanding clays, potassium ions are tightly bound in the interlayer, which prevents swelling and also makes cation exchange more difficult. (Chang, 2012) Vermiculite and smectite clays are expanding clays, while illite and muscovite are non-expanding.

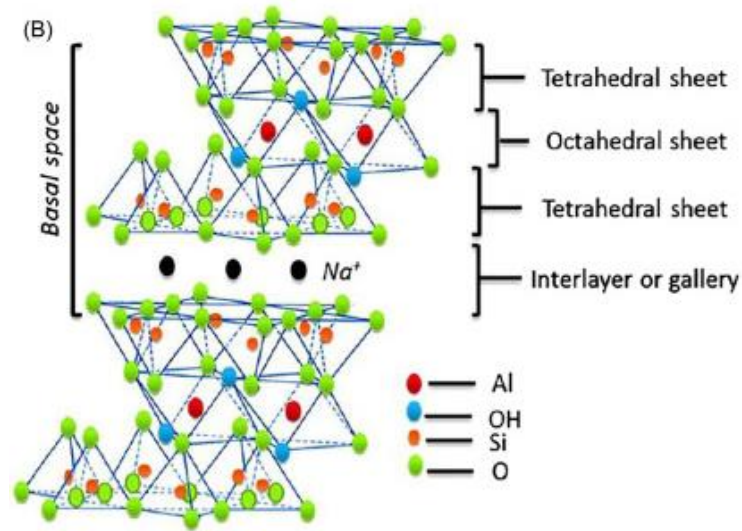


Figure 3. Example of the 2:1 layer structure in clay with octahedral alumina sheet between two tetrahedral silica sheets. (Arabmofrad et al., 2020)

Weathering of clay minerals can produce expansions on the edges of their interlayer known as a frayed edge site (FES). Frayed edge sites have larger basal spacing and are negatively charged, creating favorable conditions for Cs adsorption. (Park, 2019) Laboratory experiments using soil collected from Fukushima found that weathered biotite (WB) adsorbed Cs more effectively than other clays. (Kogure, Mukai, and Kikuchi, 2019) Similar results were obtained by Fujii et. al, (2015) on Cs uptake experiments performed on soil obtained from abandoned rice paddies within the exclusion zone. They found that Cs is primarily adsorbed in iron containing smectite and weathered biotite, and that a nonhomogeneous distribution of Cs existed in the soil. The soil's composition and ability to retain Cs presents a problem since Fukushima soils are abundant in WB and other types of 2:1 clays. (Kogure, Mukai, and Kikuchi, 2019)

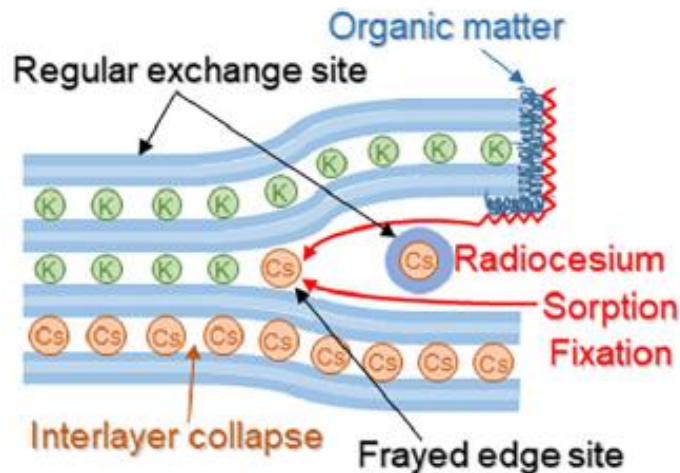


Figure 4. Example of a FES with Cs fixating into the interlayer. (Tachi et. al, 2019)

The presence of organic material affects Cs sorption and transport in the soil. Humic and fulvic acids, produced naturally from organic material, can block exchange sites on the minerals and therefore inhibit Cs adsorption by reducing the number of available exchange sites. (Park, 2019)

Removal of both particulate bound and dissolved Cs from environment is an ongoing challenge. Over the course of years following the accident, Cs has had the opportunity to migrate deeper into soil layers and farther into the interlayers of WB, making extraction difficult. Kogure, Mukai, and Kikuchi (2019) had found some success removing Cs bound to natural WB using HCl. The HCl partially dissolved the mineral's structure and leached Cs near the surface, along with other elements.

2.2 Cs Particles

Unique observations were made about the form of radioactive cesium released from the Fukushima reactor accident. Early atmospheric studies hypothesized that Cs particles released were soluble and carried in the atmosphere by sulfate particles ranging from 0.5–0.6 μm in size. (Konoplev, 2016) Samples taken at the same locations observed this as well as a spherical particulate form of Cs. During early stages of the reactor accidents, spherical particles with radioactive Cs were directly

collected on aerosol samples days after the accident on March 14th and 15th. Some of these particles included high amounts of Fe and Zn along with minor amounts of Cl, Mn, and O, and were not easily dissolvable in water. (Adachi, Kaijino, Zaizen and Igarashi, 2013) Some of these glass-like particles contained trace amounts of reactor fuel elements, were insoluble in water, and more than 2 μm in diameter. (Konoplev 2016) Studies performed by Abe et. al (2021) on Cs particles collected on March 15th, 2011 from eight sample stations located in the greater Kanto Region (over 100 km from the Fukushima Dai-ichi accident) found Cs microparticles about 1 μm in diameter with glass bodies containing less than 1 Bq per particle. Similar Cs particles, consisting of silicate glass, were found on the masks of decontamination workers near the power plant 5 years after accident. (Higaki et. al, 2017) The properties of the Cs leaching from the glass and its behavior are not well known and their long-term impact on the environment could potentially cause concern.

2.3 Preconcentration methods for Cs

Preconcentration is the process of concentrating the analyte of interest while simultaneously reducing the sample amount that needs to be processed prior to analysis. Preconcentration helps prevent trace amounts of material from going undetected and enables the analyst to achieve lower limits of detection. In this study, preconcentrating Cs also involves adsorbing the dissolved element. The process can be accomplished through contact with adsorbents, such as PB, and resins, precipitation with suitable reagents, such as ammonium molybdophosphate (AMP), and other techniques mentioned below. (Daum, 2018)

2.3.1 Tanaka, Yoshida, and Matsumoto 2015

Tanaka, Yoshida, and Matsumoto (2015) attempted to extract tightly bound Cs ions utilizing an ion exchange method involving electrophoresis. Decontaminated land was simulated by spiking Toyoura sand with known amounts of Cs. The sand was placed within a chamber and exposed to a DC field, after

being mixed with an electrolyte and topped with adsorbent material made of hydroxyapatite (HAp) and unwoven cloth. In the small chamber study, 200 g of sand was spiked with 38.1 mg of Cs and about 88% of Cs was extracted within 24 hr. In the large chamber study, 38.1 mg of Cs was added to every 100 × 100 sq. mm inside a 100 cm × 70 cm chamber. About 47% of the Cs was extracted in 48 hrs.

2.3.2 Saiki et. al 2014

Saiki et al., (2014) tested molybdophosphate type cesium adsorbent on leaching water that had collected under stacks of plant waste at temporary waste storage sites. The adsorbent was synthesized using a radiation-induced graft polymerization method. The grafting process involved immobilizing ammonium molybdophosphate onto polyethylene/polypropylene fabric by irradiating the fabric with a 2 MeV electron beam, soaking it in a monomer solution and then drying it. The batch and column tests showed a high removal efficiency, up to 75% after one batch run.

2.3.3 Kawamoto et. al 2014

Kawamoto et. al (2016) explored the possibility of decontaminating ash using nano sized PB. The study synthesized $\text{Fe}[\text{Fe}(\text{CN})_6]_{0.75}$ PB nanoparticles ranging from 8-20 nm in diameter. They fixated the nano particles onto cotton cloths to create a filter column and placed these columns within a customized pilot plant designed for contaminated ash washing. Exceptional results were achieved. One ton of contaminated combustible waste was reduced to 7.8 kg of regular waste, 6.0 kg of waste that must be disposed at managed sites and 4.4 g of waste that must be stored at interim facilities.

While multiple methods of Cs extraction showed great success, difficulties exist in obtaining the particular equipment required to produce or create specialized sorbents. Also, some extraction methods, such as using HCl, may have unwanted environmental effects. One common theme in successful extractions is the usage of Prussian blue. Soluble forms of PB can not be easily removed from solutions through filtration or centrifugation and therefore require an insoluble host to bind to that can

then be separated from the solution using physical means. With the utilization of nanodiamonds as host, the potassium hexacyanoferrate adsorbent could be an effective method to preconcentrate Cs while simultaneously providing an easy means of separating the adsorbent from the solution.

CHAPTER 3: MATERIALS & METHODS

3.1 Materials

The following materials were used in the preconcentration studies and the microparticle size categorization attempts:

- Copper chloride, Sigma-Aldrich, ACS reagent grade, Lot# SLCH2425
- Potassium ferrocyanide, Sigma-Aldrich, Lot# 0000107053
- Nanodiamonds, 0.1 wt. %, 10 nm in diameter, ADÁMAS nano, Batch No. NDMNOX10nm040821
- Nanodiamonds, 0.1 wt. %, 30 nm in diameter, ADÁMAS nano, Batch No. NDMNOX30nm040821
- Nanodiamonds, 0.1 wt. %, 50 nm in diameter, ADÁMAS nano, Batch No. NDMNOX50nm040821
- Cs-137 solution, CsCl in 0.1M HCl, 7.4 MBq/mL, Eckert & Ziegler Isotope Products, SN: 1925-2-2
- Centrifuge tubes, 15 mL
- Nalgene jars, 60 mL
- Nalgene bottles, 125 mL
- Ceramic crucibles
- Thermolyne furnace, Thermo Scientific, Model: F6000
- Centrifuge
- Coaxial HPGe Detector, P-type, 60% efficiency, EG&G ORTEC Model –S/N---
- 0.45 µm filter paper, 47 mm diameter, Omnipore
- 0.2 µm filter paper, 47 mm diameter, Omnipore
- 0.1 µm filter paper, 47 mm diameter, Omnipore
- IAEA Reference #381, Irish Seawater
- Mixed gamma verification standard, filter paper, flat, Eckert & Ziegler Analytics SRS: 78691-174

3.2 Nanodiamond Preparation

For initial sorbent testing, 10 nm, 30 nm, and 50 nm diameter nanodiamonds were obtained from ADÁMAS nano. Ten mL of a 1 wt% suspension of each were pipetted into ceramic crucibles and dried at 400°C for 8 hours in a Thermolyne furnace (Figure 5). The diamonds were then massed and diluted to 1 wt% using DI water (Figure 6). The solutions were ultrasonicated for about 30 minutes. These diluted nanodiamond solutions were then used in the adsorbent preparations.



Figure 5. Nanodiamonds after drying in the Thermolyne furnace.

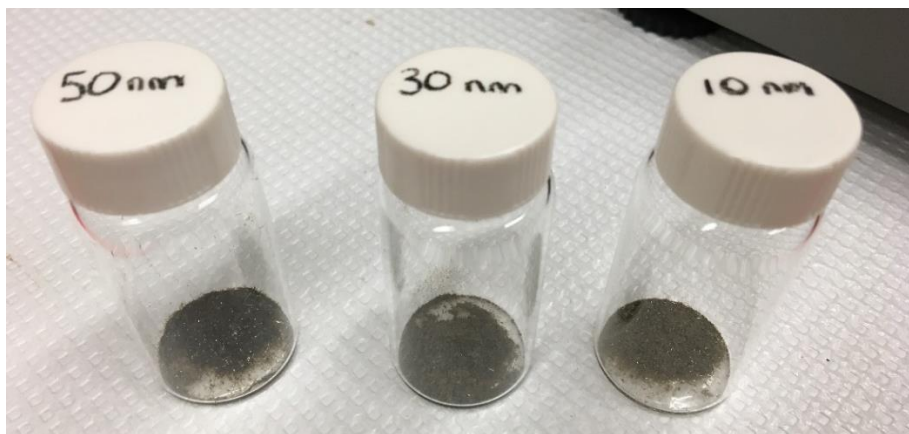


Figure 6. Nanodiamonds transferred to scintillation vials prior to dilution.

3.3 Adsorbent Preparation

The PB adsorbent was prepared by adding 0.4 mL of 1 wt% nanodiamond to 40 mL of DI water and stirring the solution for 1 minute. Subsequently, 0.4 mL of 0.1 M CuCl_2 was added to the solution and stirred for 5 minutes. Finally, 0.4 mL of 0.05 M $\text{K}_4[\text{Fe}(\text{CN})_6]$ was added to the solution and stirred for 1 hour. The solution was left overnight to precipitate. The next day, excess supernatant was pipetted off and the solutions were poured into petri dishes to let the precipitate air dry (Figure 7). The dried adsorbent was used in in the sorbent studies.

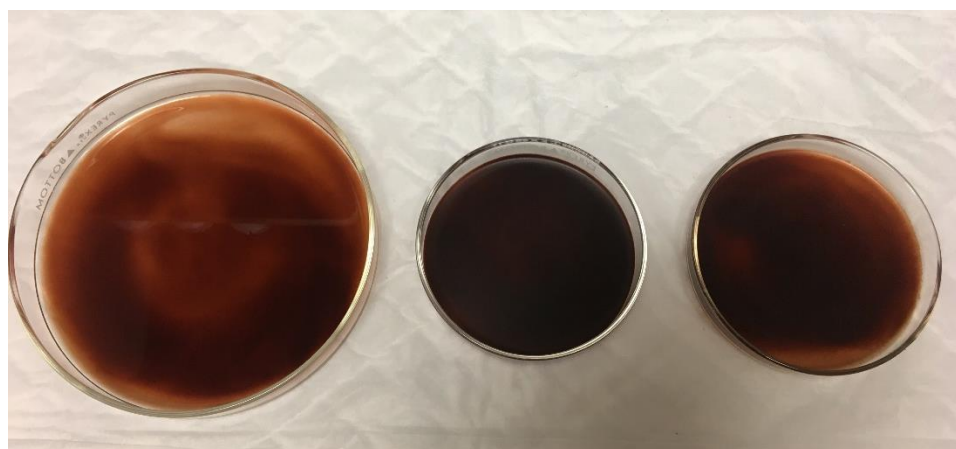


Figure 7. Nanodiamond Prussian blue adsorbent after drying.

3.4 Adsorbent Studies

Various parameters affecting the adsorption process were studied by adjusting one parameter at a time. The parameters investigated are summarized in Table 2. In each study, other than the resin concentration experiments, 15 mg of sorbent was placed in a 15 mL centrifuge tube. With the exception of the concentration study, the tube was filled with 9.9 mL of DI water and 0.1 mL of 1000 Bq/mL Cs-137 stock solution. The samples were shaken for a given time and at a given temperature as listed in Table 2 and then left to settle overnight. The next day, the samples were centrifuged for 10 min at 3000 RPM. The supernatant was poured into a 60 mL Nalgene bottle and then counted on a HPGe detector.

Table 2. Summary of the Adsorbent Studies and Parameters

Study	Sorbent Mass(mg)	DI Water (mL)	1000 Bq/mL Cs-137 Stock (mL)	Shake Time (hr)	Shake Temperature (°C)
Nanodiamond Size	15	9.90	0.10	6	Room Temp
Shake Time	15	9.90	0.10	1, 2, 3, 6, 12	Room Temp
Shake Temperature	15	9.90	0.10	6	25, 30, 35, 40, 45
Cs-137 Concentration	15	9.999, 9.99, 9.9, 9.0	0.001, 0.01, 0.1, 1.0	6	Room Temp
Resin Concentration	5, 10, 15	9.90	0.10	6	Room Temp
pH	15	9.90	0.10	6	Room Temp

A desorption study was also carried out with the 30 nm size nanodiamond adsorbent using 15 mg of sorbent and 100 Bq of Cs-137. After removing the supernatant as mentioned above, the adsorbent was resuspended in 10 mL of DI water and shaken at time intervals of 1, 6, 12, and 24 hrs. The samples were left overnight to settle and then centrifuged the next morning. The supernatant was again poured off and counted using a HPGe detector to determine the amount of Cs removed from the sorbent.

3.5 Low Activity Study

The adsorbent was tested using a reference material containing a low activity of Cs-137 to see how well it could preconcentrate Cs from a more realistic sample. The IAEA Certified Reference Material 381, Irish Seawater, (Figure 8) was used as source containing a known amount of Cs-137. A 125 mL Nalgene bottle was filled with 100 mL of the seawater. The sample was then counted for 24 hours. Afterwards, 15 mg of the PB nanodiamond adsorbent was added, and the sample was counted again for 24 hours. A comparison of the net counts under the Cs-137 photopeak was made to determine the effectiveness of the adsorbent.

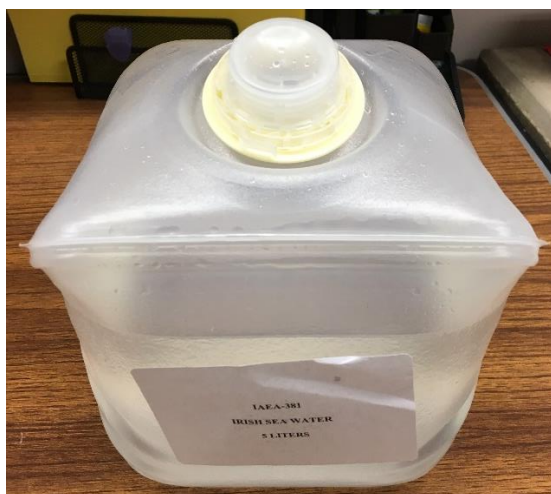


Figure 8. IAEA reference #381, Irish Seawater.

3.6 Filtration

All of the fractions from the sequential extraction study by McNabb had been retained and were stored at 4° Celsius prior to this work. Two fractions with relatively high Cs-137 activity were selected for sequential filtration. Fraction #3, the Fe/Mn oxide fraction, and fraction #5, the persistently bound fraction, from sample number F1-15-T5 were subsequently filtered through a 0.45 μm , 0.2 μm , and 0.1 μm filter. A picture of the filtration setup is shown in Figure 8. A filtration flask was connected to a

vacuum line which created a pressure difference across the filter. The filter paper sat on a fritted glass support base, which was inserted inside a rubber stopper that created a seal with the flask. A glass funnel was clamped to the support base which received the liquid. This method of filtration was performed for each size deviation, in order of largest to smallest filter pore size (Figure 10).



Figure 9. Filtration set up used to for cesium microparticle categorization.

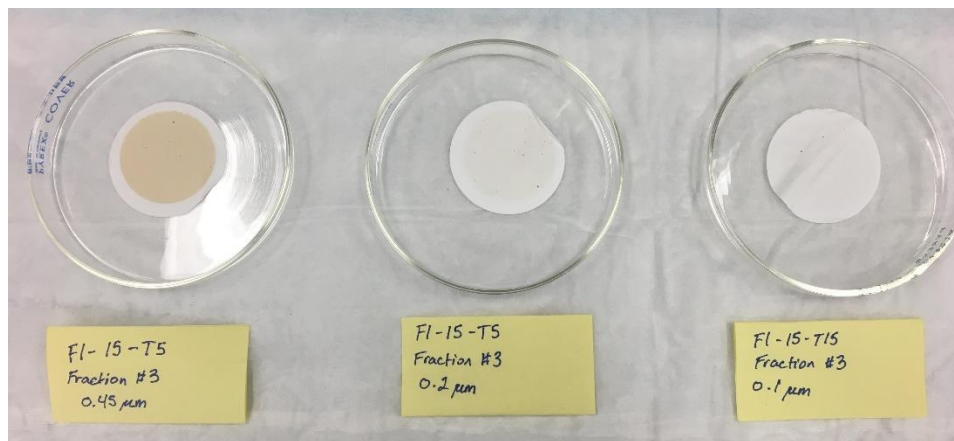


Figure 10. Filter paper after filtration of sample F1-15-T5 fraction #3.

3.7 Counting

All standards, supernatant from the sorbent studies, and the filter paper from the microparticle studies were counted for Cs-137 activity using an electrically cooled coaxial 60% efficient EG&G ORTEC Model P-type HPGe detector, laboratory designation “Curie” (Figure 11). The supernatant from the adsorbent studies along with their standards, were counted for 30 minutes, with the exception of the experiment using only 1 Bq of Cs-137. This sample was counted for 3 hours. The diameter and shake time studies were counted using a 60 mL Nalgene container, while the remainder were counted using a 125 mL Nalgene bottle of the same diameter.

The filter papers and filter paper standard were counted on a second electrically cooled coaxial 60% efficient EG&G ORTEC Model P-type HPGe detector, laboratory designation “Joliot” (Figure 12). They were counted for 24 hours on petri dishes. The filter paper standard had a Cs-137 decay corrected activity of 0.75 μ Ci with a 2.8% uncertainty.



Figure 11 & 12. EG&G ORTEC HPGe detector and lead shield used for counting the adsorbent studies.

CHAPTER 4: RESULTS

4.1 Nanodiamond Diameter Size Study

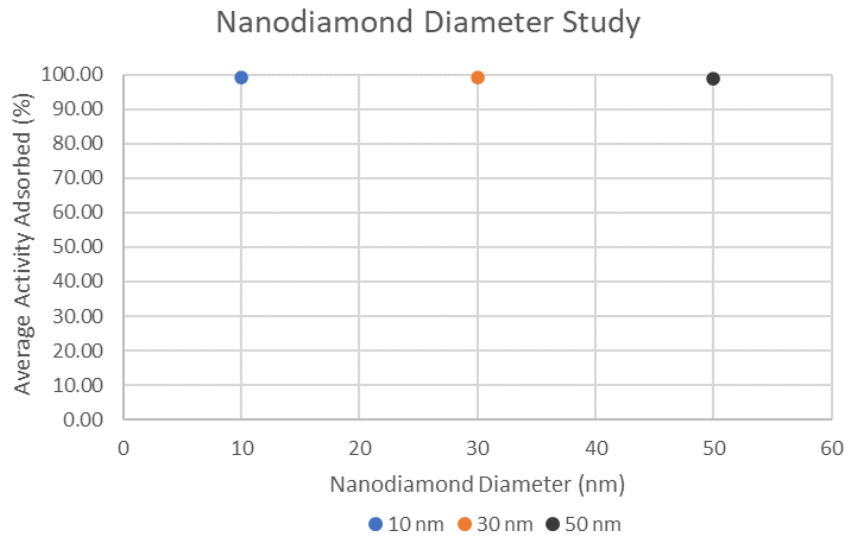


Figure 13. The average percent of activity retained by the absorbent using listed nanodiamond diameters.

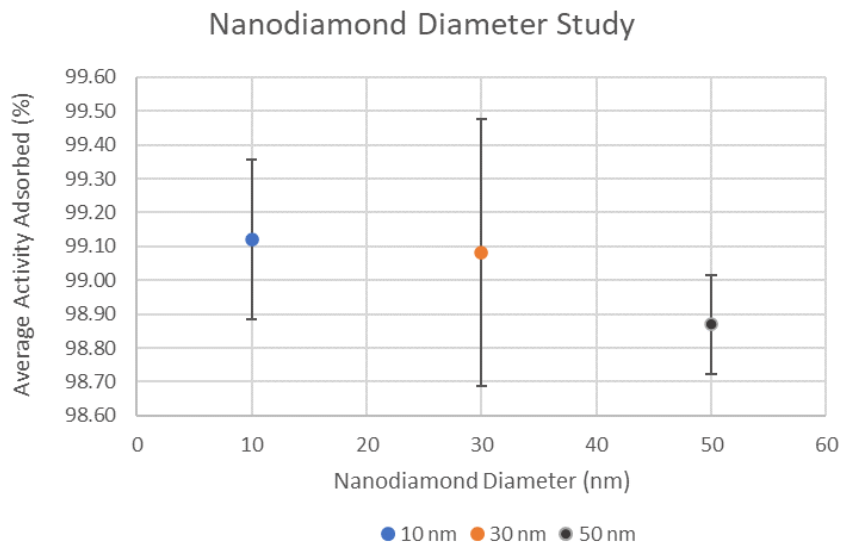


Figure 14. A close up of the average percent of activity adsorbed at different nanodiamond diameters.

The 10 nm, 30 nm, and 50 nm diameter nanodiamond sorbents were compared to each other to identify which nanodiamond size best retained Cs-137 particles. All three sorbents performed exceptionally well, adsorbing approximately 99% of the cesium. The 10 nm, 30 nm, and 50 nm nanodiamond accumulated on average 99.1%, 99.1%, and 98.9% of activity, respectively (Figure 13 and 14). The 30 nm diamonds adsorbed approximately the same amount of Cs and seemed to perform better than the 10 nm and was selected for the subsequent studies.

4.2 Contact Time Study

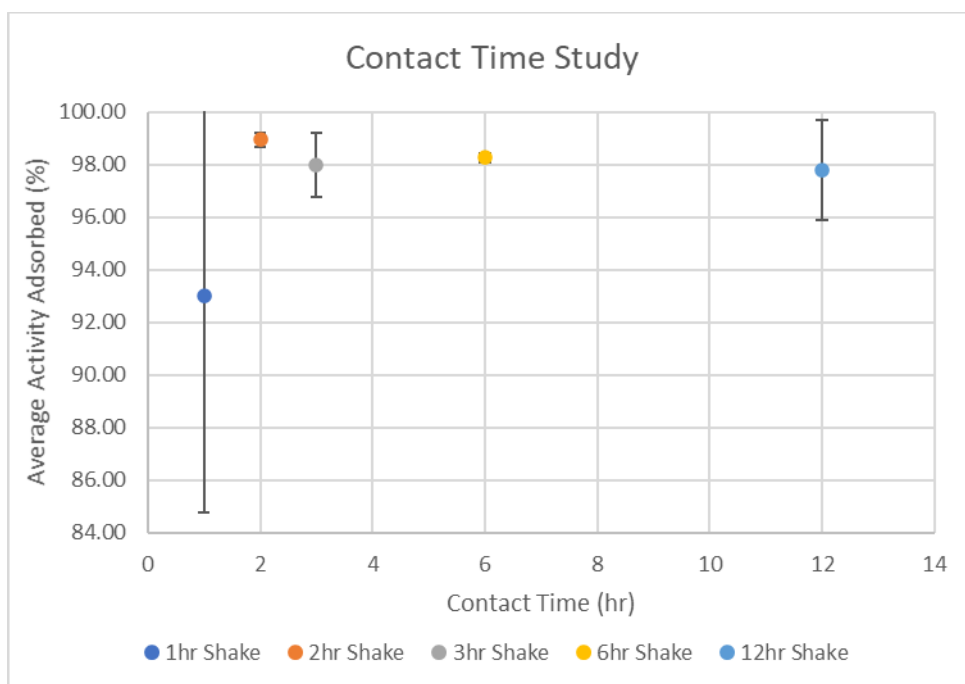


Figure 15. The average percent of activity retained by the absorbent during different contact times.

The shaking time of each sample set was varied in order to determine if contact time affected Cs adsorption. The 1 hour shaking time resulted in the lowest amount of activity adsorbed (93.0%, Figure 15). Shaking times of 2 hours and longer performed better, with a difference of about 2.2%. After the 2-hour mark, some of the Cs might become desorbed again from sorbent, indicating possible progress towards an equilibrium within the solution. A similar trend is observed in the desorption study.

4.3 Temperature Study

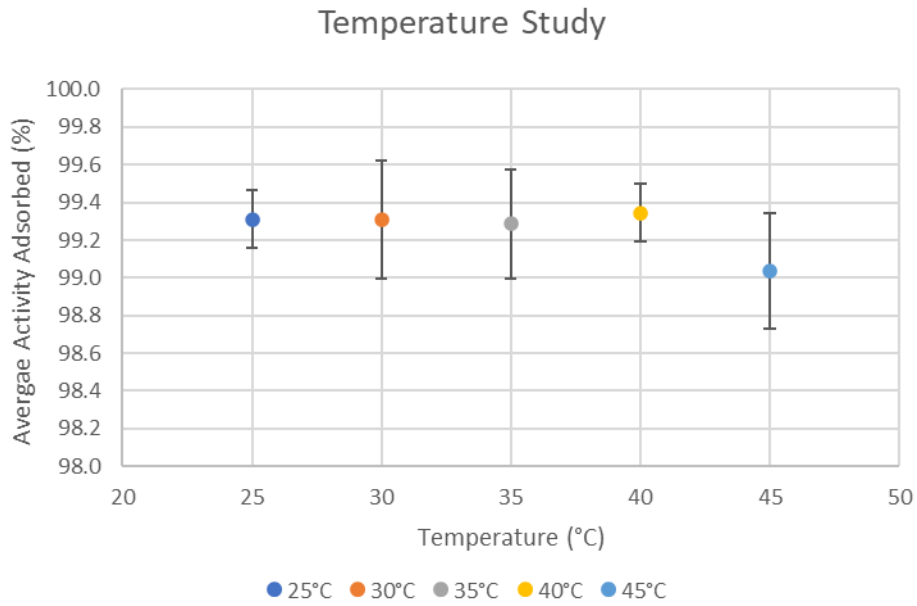


Figure 16. The average percent of activity retained by the absorbent at listed temperatures.

The temperature at which the sample were shaken was varied in order to determine if there is a thermodynamic effect on the Cs adsorption (Figure 16). Between 25 to 40°C, the adsorption percentage was consistent at approximately 99.3% and then started to drop as the temperature increased above 40°C.

4.4 Cs Concentration Study

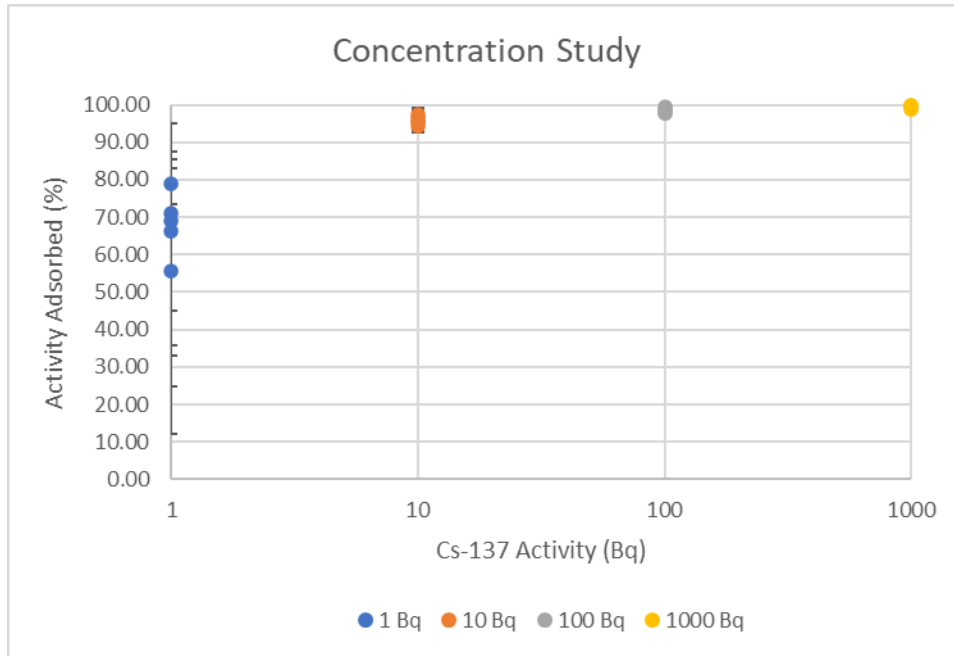


Figure 17. The percent of activity retained by the adsorbent at listed Cs-137 activities.

The amount of Cs added to each sample set was varied in order to determine the sensitivity of the adsorbent (Figure 17). The average amount of activity adsorbed in the 1 Bq, 10 Bq, 100 Bq, and 1000 Bq, samples were 68.2%, 95.9%, 98.5%, and 99.6%, respectively. The adsorbent did not seem to perform as effectively at the lowest activity concentration. All samples were counted for 30 minutes initially. The 1 Bq samples were recounted for three hours in order to reduce the uncertainty associated with the measurement. Even with the longer counting times, the 1 Bq samples still produced a large error.

4.5 Adsorbent Concentration Study

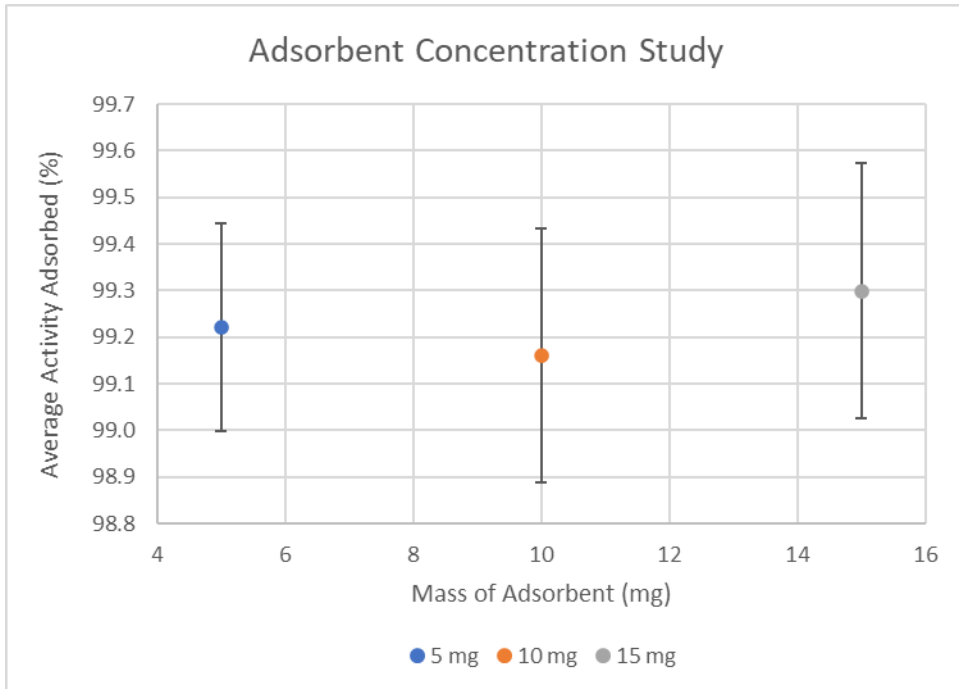


Figure 18. The percent of activity retained by the adsorbent at listed masses.

Specified amounts of adsorbent were added to each sample set in order to analyze how mass affected retention (Figure 18). The sample results were very consistent, with only about a 0.2% difference in Cs-137 adsorption. A mass of 5 mg of adsorbent was sufficient to achieve an extraction greater than 99%. The samples containing 15 mg of adsorbent retained the most Cs, leaving behind only 0.70 %.

4.6 pH Study

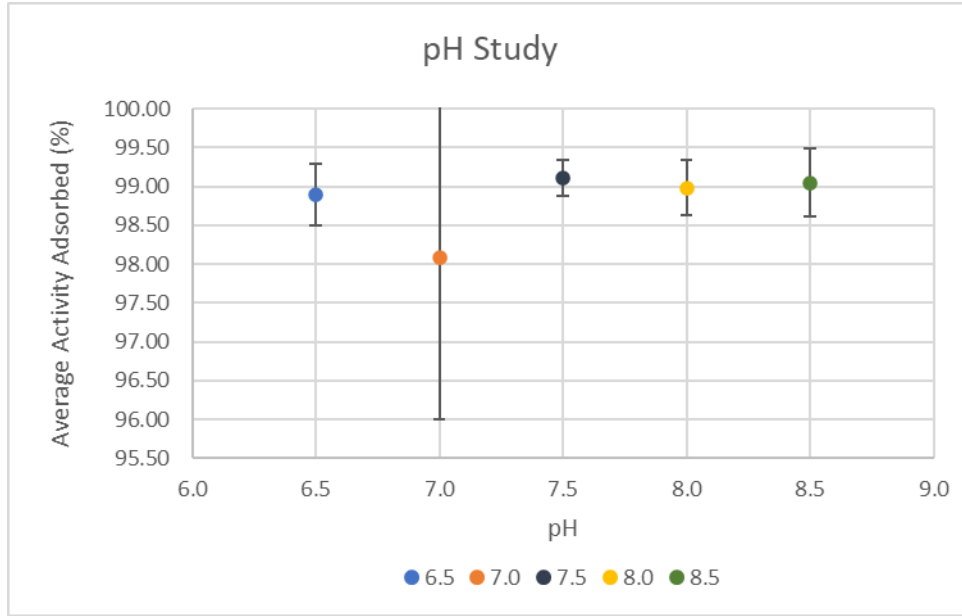


Figure 19. The percent of activity retained by the adsorbent at listed pH values.

The pH of the DI water added to each sample set was adjusted to a predetermined value in order to observe adsorbent response. The sorbent retention did not change considerably within a pH range of 6.5 - 8.5 (Figure 19). There is a slight drop in the 7.0 efficiency due to one sample reading higher than the other. This contributed to the lower performance of the 7.0 group as well as a higher error.

4.7 Desorption Study

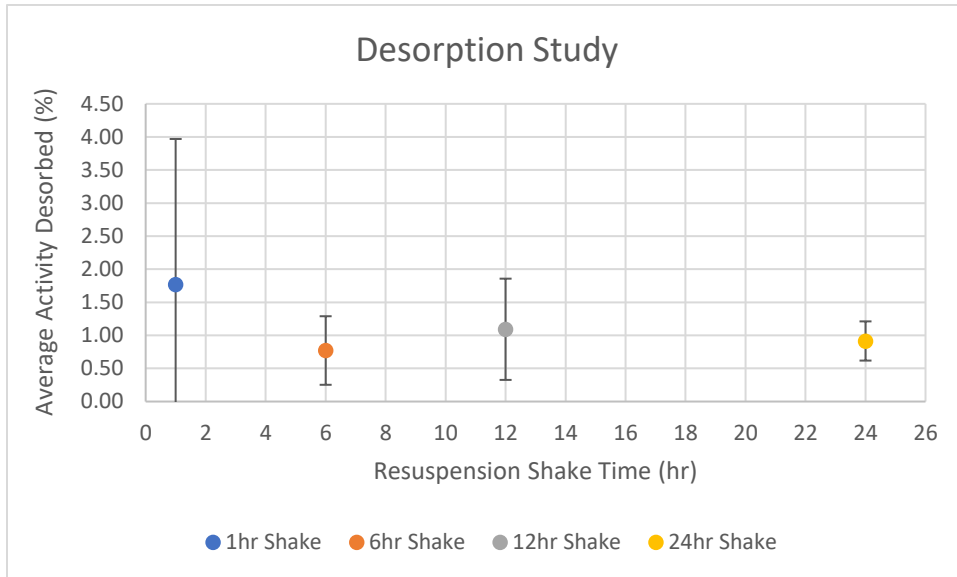


Figure 20. The percent of activity resuspended in the water following listed shake times.

The adsorbent's ability to retain Cs over longer periods of time was tested by resuspending and shaking the adsorbent in pure DI water. Resuspension times of 1 hr, 6 hr, 12 hr, and 24 hr were investigated (Figure 20). The largest release of Cs was seen in the 1 hr sample, with an average of 1.8% of the activity being released back into the solution. At the other shaking times, the range of activity released was between 0.8% to 1.1%. The release of Cs from the adsorbent may be due the sample progressing towards an equilibrium with the DI water as indicated by the slightly fluctuating trends with increasing resuspension times.

4.8 Low Activity Study

100 mL of IAEA reference Irish Seawater was counted before and after adding adsorbent and the pictures of their respective gamma spectras are shown in Figures 21 and 22.

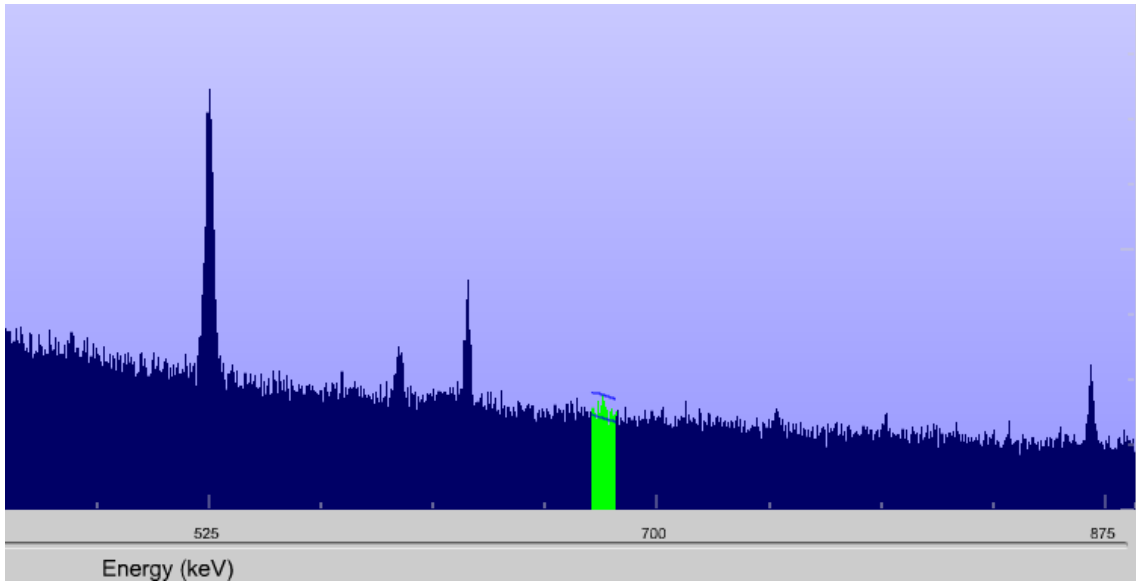


Figure 21. Gamma spectra for Cs-137 photopeak before adding adsorbent.

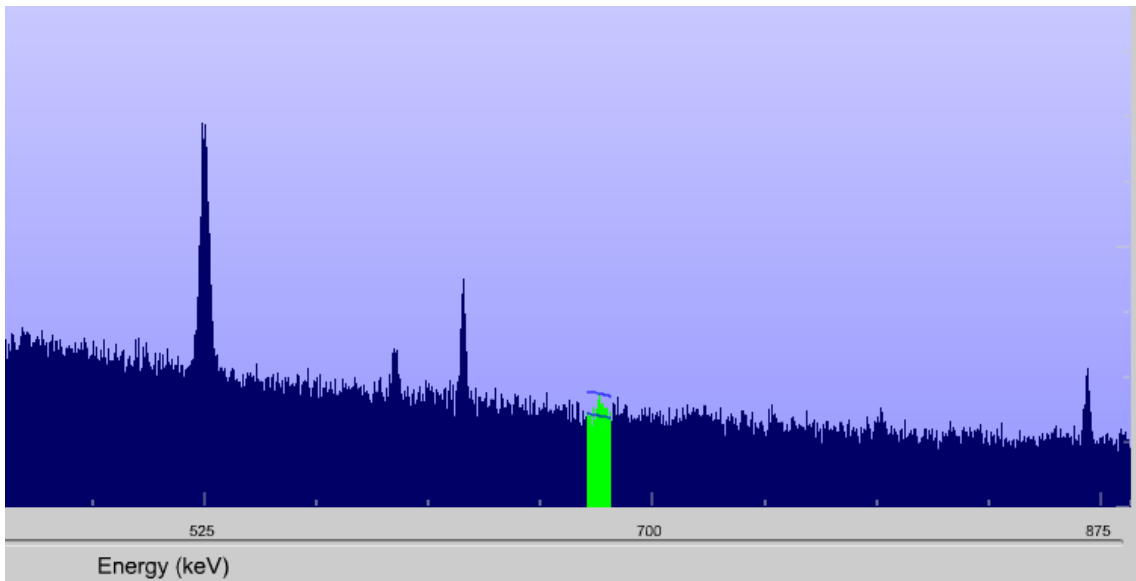


Figure 22. Gamma spectra for Cs-137 photopeak after adding adsorbent.

Visually there is no change in the Cs-137 photopeak. The net counts under the photopeak of only seawater were 258 ± 16.1 counts. The net counts after adding the adsorbent were 165 ± 12.8 counts. There was about a 63% decrease in net counts after adding the adsorbent.

4.9 Cs Particles

The Cs size categorizations were performed by filtering previously collected soil samples that have undergone sequential extraction. The amount of activity within each size range was determined by comparing the total counts from the filter paper to a manufactured standard. The results are shown in Figures 23 and 24.

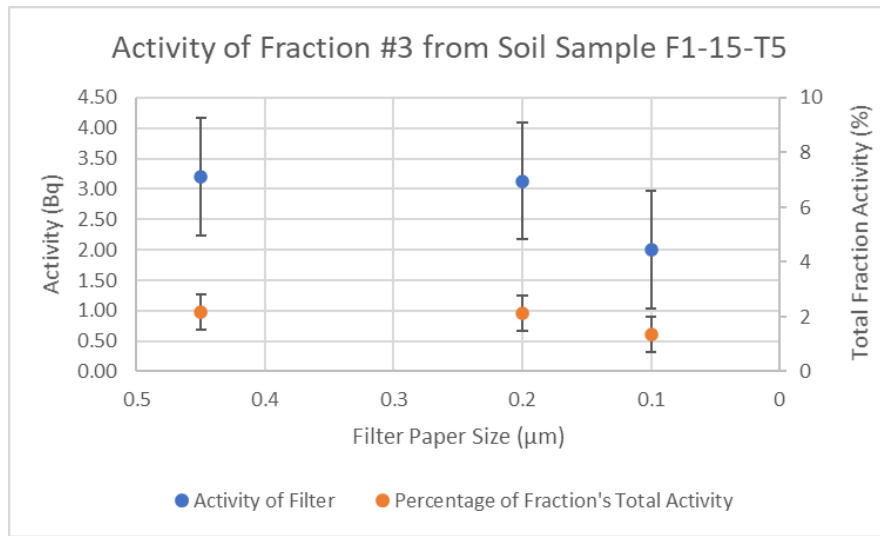


Figure 23. Cs-137 activity measured on each filter after filtration of fraction #3.

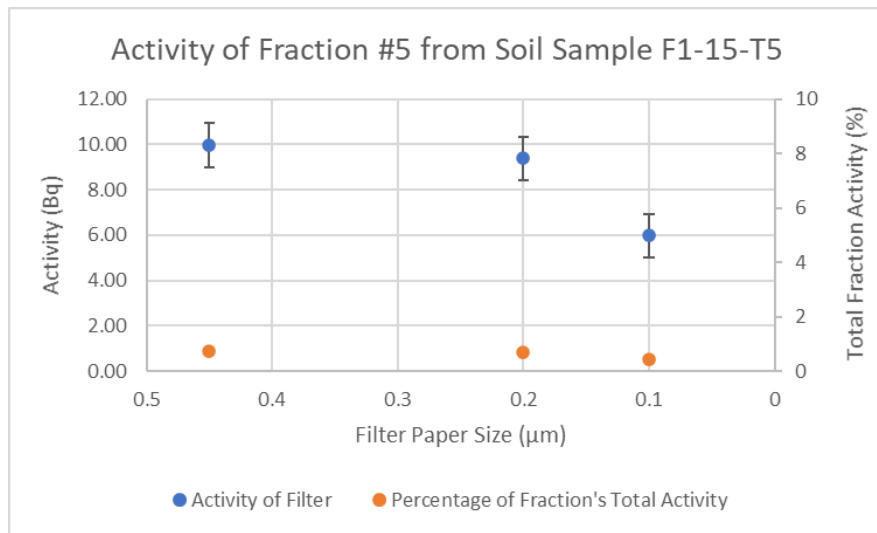


Figure 24. Cs-137 activity measured on each filter after filtration of fraction #5.

The Cs-137 activities retained by the 0.45 μm , 0.2 μm and 0.1 μm filters after filtration of fraction #3 were 3.2 Bq, 3.1 Bq, and 2.0 Bq, respectively. The total amount of Cs-137 previously found in fraction #3 before filtration was 147 Bq. This would indicate that 2.2% of RCs is associated with a size fraction $\geq 0.45 \mu\text{m}$, 2.1% is associated with a size fraction between 0.45 - 0.2 μm , and that 1.4% is associated with a size fraction between 0.2 μm - 0.1 μm . Overall, only 5.7% of the total activity of fraction #3 was retained by the filters. The majority of the Cs-137 present in fraction #3 is therefore believed to be in dissolved form.

The Cs-137 activity present in fraction #5 was almost three times the amount present in fraction #3. The activities found on the 0.45 μm , 0.2 μm and 0.1 μm filters were 10 Bq, 9.4 Bq, and 6.0 Bq, respectively while the overall activity previously determined for this fraction was 1335 Bq. The filtration results for fraction #5 would indicate that 0.7% of Cs is associated with a size fraction $\geq 0.45 \mu\text{m}$, 0.7% is associated with a size fraction between 0.45 - 0.2 μm , and that 0.4% is associated with a size fraction between 0.2 μm - 0.1 μm . Overall, only 1.8% of the total activity of fraction #5 was retained by the filters. The majority of the Cs-137 present in fraction #5 is therefore believed to be in dissolved form.

CHAPTER 5: DISCUSSION

5.1 Adsorption Studies

The nanodiamond diameter study determined the optimal size of nanodiamonds to be used as a substrate for Cs-137 preconcentration studies. The 30 nm diamonds adsorbed approximately the same amount of Cs as the 50 nm but seemed to perform more consistently than the 10 nm. The larger error and slightly lower performance from the 30 nm samples may have been due to a possible outlier in the data set. Overall, the 30 nm nanodiamonds were chosen for the subsequent preconcentration studies.

The diameter study was also consistent with results obtained in batch studies performed by Matsumoto et al. (2018). Their studies tested the efficiency of detonation nanodiamond (DND) Cu-PB adsorbent to adsorb Cs-137 from deionized and seawater samples. They reported an over 99% efficiency in Cs adsorption in artificial seawater batch additions. (Matsumoto, 2018) The sorbent also performed similarly to a manufactured sorbent called ANFEZH, a cellulose grain coated by double iron-potassium ferrocyanide that has been reported to have a 95–98% Cs removal efficiency. (Remez, 1998)

For the mass study, all the 5 mg, 10 mg, and 15 mg samples performed very similarly with 99.2%, 99.2% and 99.3% retained, respectively. As little as 5 mg of sorbent was needed to effectively adsorb 1000 Bq of Cs-137 from a 10 mL sample. These results show that small amounts of material are sufficient to concentrate large amounts of Cs. The adsorbent did seem to have a harder time capturing Cs in the 1 Bq samples. There could be a reduced performance at low activities due to how little Cs atoms are available in the sample for adsorption. Also, the errors from sample geometry and counting play a larger role in adsorbent's efficiency. Further investigation into mass and concentration ratios could be explored to find the point when performance decreases.

Based on the trends from shake time and desorption study, the cesium retained by the sorbent appears to have established an equilibrium with the solution after 2 hours. The apparent establishment of a chemical equilibrium demonstrates that RCs is relatively rapidly adsorbed under mixing conditions. Application of this sorbent can save time and quickly process contaminated water.

The variation of pH did not seem to alter sorbent performance, with the exception of pH7. During the preparation of the pH7 batch, a part of the sorbent may have been transferred together with the supernatant, resulting in a much higher activity than the in the other samples tested at a pH of 7.0. This would explain the resulting slight dip in adsorption. The sorbent should perform as expected at this pH.

Due to the laboratory setting in which these studies were performed, it is not known if the sorbent performance will be equivalent in the field and operational situations. Therefore, further experiments are recommended to test the sorbent in solutions with competing cations such as potassium, and sodium. Also tests with artificial seawater and brackish waters spiked with known amounts of RCs are also recommended. Further ranges of temperatures can be explored since oceanwater can be below freezing depending on location.

5.2 Low Activity Study

An unexpected drop in net counts occurred after adding adsorbent to the seawater sample. It is unclear why such a large drop occurred. If the adsorbent was unsuccessful at concentrating the Cs, the adsorbent sample should have produced about the same number of counts as the seawater only sample. In the Cs concentration study, the 1 Bq sample exhibited reduced efficiency compared to the higher activity samples. Due to the low Cs-137 activity present in the seawater, the sorbent may not have performed as well. Also, there is no information available on the amount of stable Cs present in the water. Plus, the seawater was acidified to a pH of 1 and it is not known if the sorbent could work in this

acidity. Further investigation is needed to conclude if the adsorbent is unsuccessful at preconcentrating RCs in very low activity samples.

5.3 Size Categorization

The extracted fractions chosen to undergo filtration were fraction #3, the Mn/Fe oxide, and fraction #5, the persistently bound phase, from soil sample F1-15-T5 because they contained a greater amount of extracted RCs than the other soil samples. The persistently bound phase was of interest since this phase most closely represents the Cs that would have been extracted from tightly bound clays and FES. Meanwhile, the Mn/Fe oxide phase was of interest due to its high activity that was not in seen in other samples. If time had not been a factor, then all remaining fractions would have been filtered and counted.

Based on the results of the fraction #3, very little activity is seen on the filter papers. There is a slight increase in activity on the 0.45 μm and 0.2 μm filters. It is likely that a tiny amount of particulate bound Cs was extracted from the soil, however the vast majority of the Cs in fraction #3 is believed to be dissolved.

Higher Cs-137 activities were observed on each filter collected from fraction #5 compared to the corresponding filters from fraction #3. However, the overall Cs-137 activity of fraction #5 is also greater than that of fraction #3. The proportion of the overall activity of filters in fraction #5 compared to the overall activity of the fraction were very small, about 1% or less. It is likely that the small amounts of Cs-137 found on each filter collected from fraction #5 is probably due to the Cs-137 containing solution evaporating on the filter. There is a chance that a tiny amount of particulate bound Cs was extracted from the soil, however the vast majority of the Cs in this fraction is believed to be dissolved.

Based on the Cs-137 activity collected on the filter papers, it is hypothesized that sequential extraction was effective at only extracting soluble forms of Cs. Particulate bound Cs or special forms of

Cs, such as the glass beads mentioned earlier, may exist in the soil. Imaging the soil through a scanning electron microscope or x-ray diffraction analysis are recommended to verify the state of the remaining RCs.

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APPENDIX I: RAW DATA

Table 3. Nanodiamond Diameter Size Study Data

Nanodiamond Size (nm)	Average Activity Adsorbed (%)	Standard Deviation (%)
10	99.1	0.24
30	99.1	0.39
50	98.9	0.15

Table 4. Contact Time Study Data

Contact Time (hr)	Average Activity Adsorbed (%)	Standard Deviation (%)
1	93.0	8.21
2	99.0	0.26
3	98.0	1.24
6	98.3	0.17
12	97.8	1.90

Table 5. Temperature Study Data

Temperature (°C)	Average Activity Adsorbed (%)	Standard Deviation (%)
25	99.3	0.15
30	99.3	0.31
35	99.3	0.29
40	99.4	0.16
45	99.0	0.31

Table 6. Cs Concentration Study Data

Cs-137 Concentration (Bq)	Activity Adsorbed (%)	Standard Deviation (%)	Cs-137 Concentration (Bq)	Activity Adsorbed (%)	Standard Deviation (%)
1 A	69.0	16.6	100 A	97.9	0.29
1 B	79.0	16.0	100 B	98.5	0.27
1 C	66.3	16.7	100 C	99.0	0.25
1 D	55.8	17.7	100 D	97.7	0.29
1 E	71.1	16.4	100 E	99.5	0.22
10 A	95.2	1.88	1000 A	99.6	0.02
10 B	97.5	1.69	1000 B	99.6	0.02
10 C	96.7	1.72	1000 C	98.8	0.04

10 D	94.4	1,87	1000 D	99.9	0.02
10 E	95.8	1.76	1000 E	99.8	0.02

Table 7. Cs Concentration Study Data

Cs-137 Concentration (Bq)	Average Activity Adsorbed (%)	Standard Deviation (%)
1	68.2	8.39
10	95.9	1.24
100	98.5	0.75
1000	99.6	0.42

Table 8. Adsorbent Concentration Study Data

Adsorbent Concentration (mg)	Average Activity Adsorbed (%)	Standard Deviation (%)
5	99.2	0.22
10	99.2	0.27
15	99.3	0.27

Table 9. pH Study Data

pH	Average Activity Adsorbed (%)	Standard Deviation (%)
6.5	98.9	0.39
7.0	98.1	2.10
7.5	99.1	0.23
8.0	99.0	0.35
8.5	99.1	0.44

Table 10. Desorption Study Data

Shake Time (hr)	Average Activity Desorbed (%)	Standard Deviation (%)
1	1.77	2.20
6	0.77	0.52
12	1.09	0.77
24	0.92	0.30

APPENDIX II: SPECIAL PERMISSIONS

Figure 3: Reprinted from Handbook of Food Nanotechnology, 1, Sara Arabmofrad, Mahsa Bagheri, Hamid Rajabi, and Seid Mahdi Jafari , Nanoadsorbents and nanoporous materials for the food industry, 107-159, Copyright 2020, with permission from Elsevier.

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