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Spallation Neutron Source Carbon Adsorption Tests Final Report

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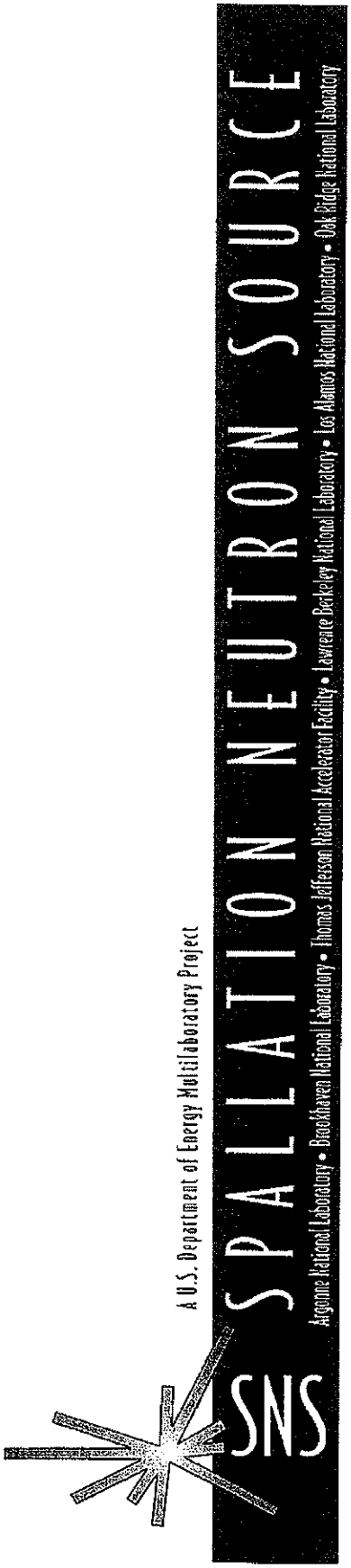

Executive Summary

In May 2001, Knight Engineering (now M+W Zander) contracted ADA Technologies, Inc. to support the design of the Spallation Neutron Source (SNS) project. SNS is a particle accelerator facility that is being constructed by the Department of Energy to conduct material science and other experiments. The facility uses an elemental mercury target to generate neutrons for these experiments. In servicing this target small quantities of elemental mercury are expected to vaporize and accumulate in the maintenance hot cell. The mercury vapor concentration will be controlled by ventilating the cell, and mercury will be removed from the ventilation exhaust by using a mercury filter system. The SNS project team evaluated several mercury control technologies and determined that the most economical option for mercury control uses sorption beds of sulfur-impregnated activated carbon. ADA has researched the mercury capacities of several of these carbon sorbents.

Mercury sorption capacities for various carbon materials were determined through controlled laboratory tests at ADA's facilities in Littleton, CO. The test matrix called for six sorption tests that examined the effects of gas velocity, carbon manufacturer, carbon particle size, and humidity of the test gas on the mercury sorption capacity. Carbon samples were requested from two suppliers for use in these tests. Nucon International Inc. submitted two samples of its MERSORB^{7} carbons. The MERSORB^{7} carbons were both extruded cylinders of different diameters (1.5 and 3 millimeters). The other carbon samples were Type HGR carbons manufactured by Calgon Carbon Corporation. The shape of the Type HGR carbons was different, with one being granular and the other cylindrical.

Test conditions for the sorption tests were chosen based on the operating conditions expected for the Spallation Neutron Source (SNS) target cell. All sorption tests were conducted at a temperature of 100°F, and a pressure of 14.3 psia with a gas stream having a mercury concentration of 50 $\mu g/m^3$. The breakthrough of mercury was monitored at various depths in the carbon beds using a combination of atomic fluorescence and atomic absorption analytical techniques. Tests were concluded when the mercury concentration exceeded 0.1 $\mu g/m^3$ at a specified sample location for a 1-second residence time. Experimental data was then fitted to the Rosen fixed-bed sorption model to calculate the mercury sorption capacities for carbons evaluated in the six sorption tests.

The majority of the sorption tests were conducted with an empty bed gas velocity of 30 feet per minute for a test gas with 25% relative humidity (RH). These conditions were used to examine the sorption capacity for carbons with different particle size and to compare the performance of the different carbons. The 1.5mm and 3mm Nucon carbons had nearly the same mercury capacity when tested with a gas at 25% RH. After normalizing the data to the same residence time, the 1.5mm carbon had better performance compared to the 3mm carbon, which is consistent with data published by Nucon. Test data also showed that the mercury capacities for the Nucon 1.5mm and the Calgon Type HGR carbons were nearly the same, with the Nucon carbon having a slightly higher capacity. A single test was conducted at a lower gas velocity of 15 feet per minute to test the effect of gas flow rate on mercury capacity. The mercury capacity for the Nucon 1.5mm carbon was greater in the test conducted at gas velocity of 15 feet per minute compared to the test run at 30 feet per minute, due to the broadening of the breakthrough curve at the higher flow rate. By far the parameter that affected the mercury sorption

capacity the most was the humidity level in the test gas. In the test conducted to measure the effect of relative humidit, the mercury sorption capacity was nearly 125% greater in the high humidity test (50% RH) compared to the capacity determined at the low humidity level (25% RH).

Test results were also used to estimate the lifetime of carbon used in a full-scale mercury filter system operating at 15 feet per minute. The carbon changed out time was calculated based on an outlet mercury concentration from the carbon filters of 0.1 $\mu g/m^3$.

Overall, the Calgon Type HGR carbon exhibited the shortest predicted lifetime of 260 days, while the longest lifetime was 900 days using the Nucon 1.5mm carbon exposed to 50% RH.

Introduction

The Department of Energy (DOE) is constructing a world-class subatomic particle generator that will provide a source of energetic neutrons for scientific research. Neutrons and other spallation products will be generated by bombarding a target of elemental mercury with an intense beam of protons. During the process, the mercury target will absorb energy from the protons and as a result, the temperature of the mercury target will increase. Estimated mercury release from the mercury target is 4.7 grams of mercury per day, resulting in a steady state mercury concentration of 43 $\mu g/Nm^3$ in the hot cell of the Spallation Neutron Source (SNS) building. The mercury concentration in the hot cell space will be controlled by circulating clean, mercury-free air through the target cell. The circulated air will be passed through a mercury filter system to remove the vapor-phase mercury prior to releasing the air to the atmosphere. Outside air will be brought into the hot cell as make-up air at a rate of 3,000 cubic feet per minute (CFM) during normal operations to maintain mercury concentrations in the hot cell to less than 50 $\mu g/Nm^3$.

The SNS project team evaluated several mercury control technologies for use on the project and decided that activated carbon bed filtration is the most cost-effective method for controlling mercury emissions in the SNS building. Activated carbon is widely used in industrial applications as an adsorbent medium for mercury and organic emission control. Carbon provides a high surface area (400 to 600 m^2/g) to facilitate physical sorption of contaminants from gas and liquid streams. Although activated carbon is an effective sorbent for elemental mercury vapors, sorption capacity is enhanced by impregnating the carbon with either sulfur or iodine. For this project, sulfur-impregnated carbons from Nucon International Inc. and Calgon Carbon Corporation were evaluated.

The SNS design requires that the outlet mercury concentration from the mercury filter be less than 0.1 $\mu g/m^3$. When the mercury concentration in the air exiting from the mercury filter exceeds this criterion, the carbon bed will be replaced with fresh carbon. Literature from carbon manufacturers indicates that chemical-impregnated carbon, under proper operating conditions, can reduce elemental mercury concentrations to less than 0.001 $\mu g/m^3$ (Calgon, 1997). However, proper operating conditions include low moisture content in the air and low gas temperatures (70°F). Air in the hot cell is expected to have moderately high relative humidity at times (25-50%) and air temperatures approaching

100°F. Laboratory tests were conducted at ADA Technologies, Inc. to confirm the mercury loading claims made by the carbon manufacturers.

In the proposed mercury filter configuration, carbon canisters will have an annular design in which the inlet gas enters the bottom of the sorbent vessel and passes outwardly through the carbon bed in a radial fashion. The treated gas is then collected at the outside diameter of the carbon bed and discharged through the top of the sorbent vessel. The hot cell gas will be split and processed through several carbon canisters connected in parallel to minimize the pressure drop of the carbon filter system. The maximum flow through a single carbon canister is 900 CFM with a gas-carbon contact time on the order of 2.5 seconds and a pressure drop of 2 inches water column across the carbon filter system.

ADA Technologies, Inc. was contracted by Knight Engineering (now M+W Zander) to determine the mercury absorption capacity for two chemically-impregnated activated carbons. Conditions for the laboratory carbon tests were intended to be similar to conditions realistic for the SNS project. Results from these tests were used to predict the change out times for various carbon adsorbents as a function of sorbent particle size, relative humidity of the gas, and gas flow rate through the sorbent bed. The optimal sorbent being one that has the maximum mercury sorption capacity and minimizes the number of carbon change outs for the life time of the SNS facility.

An alternate sorbent produced by ADA Technologies Inc. was included in the original test matrix. The ADA sorbent is a regenerable noble-metal sorbent that amalgamates with vapor-phase mercury. Using this type of sorbent would eliminate frequent carbon change out that exposes workers to radiation and would reduce the physical size of the mercury filter system compared to a carbon-based system. However, due to high capital cost of the system, this sorbent material was eliminated from consideration and was not tested.

The project was divided into four tasks to complete the scope of work identified by the SNS project team. The tasks are as follows:

Task 1 – Carbon Survey

Task 2 – Laboratory Sorbent Testing

Task 3 – Analysis of Laboratory Data

Task 4 – Project Management

In the first task ADA contacted several carbon manufactures to collect information on various chemical-impregnated activated carbons. Based on a review of the compiled data, the SNS project team considered Calgon and Nucon as the viable carbon suppliers for a project of this magnitude. Information on each of the carbon materials was collected from the carbon suppliers and presented in tabular form to allow a direct comparison of the carbon products, including unit cost. Because Nucon has experience in radiological applications, the Nucon MERSORB[7] carbon was selected as the primary carbon for the sorption tests with Calgon as the alternate carbon. Approximately 10 pounds of each carbon was obtained from suppliers for the laboratory tests.

In Task 2, ADA designed and constructed the sorption columns for the laboratory tests. Laboratory tests were conducted in a gas stream representative of conditions expected for the SNS target hot cell with regards to vapor-phase mercury concentration,

gas temperature, pressure, and moisture content. The test matrix identified seven cases to evaluate Nucon and Calgon carbons as a function of test gas humidity level and superficial gas flow rates. In all tests the sorption columns were maintained at a constant temperature of 100 °F. The sorption columns were pressurized to 14.3 psia to simulate the atmospheric pressure conditions in eastern Tennessee.

Data from the laboratory sorption tests were evaluated in Task 3. Sorption capacities (K_D) of the carbons were determined by modeling the mercury breakthrough data using the Rosen model (Rosen, 1952). With the K_D values, the time to reach a breakthrough mercury concentration of 0.1 $\mu g/m^3$ was determined as a function of bed depth for each carbon test. The Bed Depth Service Time model (Cooney, 1999) was used to predict the lifetime for carbon in a full-scale mercury filter system. For our purposes, the Bed Depth Service Time model was used to predict when the mercury concentration in the outlet gas from the carbon bed would reach 0.1 $\mu g/m^3$.

In Task 4 the test plan and final report for the project were prepared.

Description of Carbon Sorbent Materials

Carbon sorbent samples were obtained from Nucon International and Calgon Carbon Corporation for the laboratory tests. All carbons were sulfur-impregnated with roughly 10 to 18% sulfur by weight. Nucon provided two carbon samples, both of them cylindrical extrudates (pellets). The average diameters of the carbon pellets were 1.5mm and 3 mm, with a length ranging from 6 to 10 mm. Equivalent spherical diameters were found to be 2.3 mm and 4.1 mm, respectively. Calgon also sent two carbon samples for testing. The first was a granular Type HGR carbon with a particle diameter ranging from 1.7 to 4.8 mm. The average particle diameter for the Type HGR carbon was found to be 2.7 mm. The second carbon from Calgon was a pelletized carbon with a cylindrical shape, denoted as Type HGR-P. The pellet material had a particle diameter of 4 mm and a length of 6 mm. Table 1 summarized the properties of each of the carbon sorbents.

Table 1. – Summary of Carbon Properties

	Calgon Carbon Corp.		Nucon International		
Type	HGR	HGR-P	MERSORB	MERSORB	MERSORB
Shape	granular	pellet	pellet	pellet	Pellet
Diameter (mm)	1.7 - 4.8	4	4	3	1.5
Length (mm)	n/a	6	6-10	6-10	6-10
Density (g/cc)	0.6	0.6	0.53 - 0.6	0.53 - 0.6	<0.53
Sulfur Content (%)	10 - 18	10 - 18	10 - 15	10 - 15	10 - 15

Description of Test Equipment

The test fixture used in the sorption tests is shown in Figure 1. The photo shows the two sorption test chambers, humidification chamber, hot air heater, mass flow controllers, and temperature controllers. A description of the equipment used on this project follows.

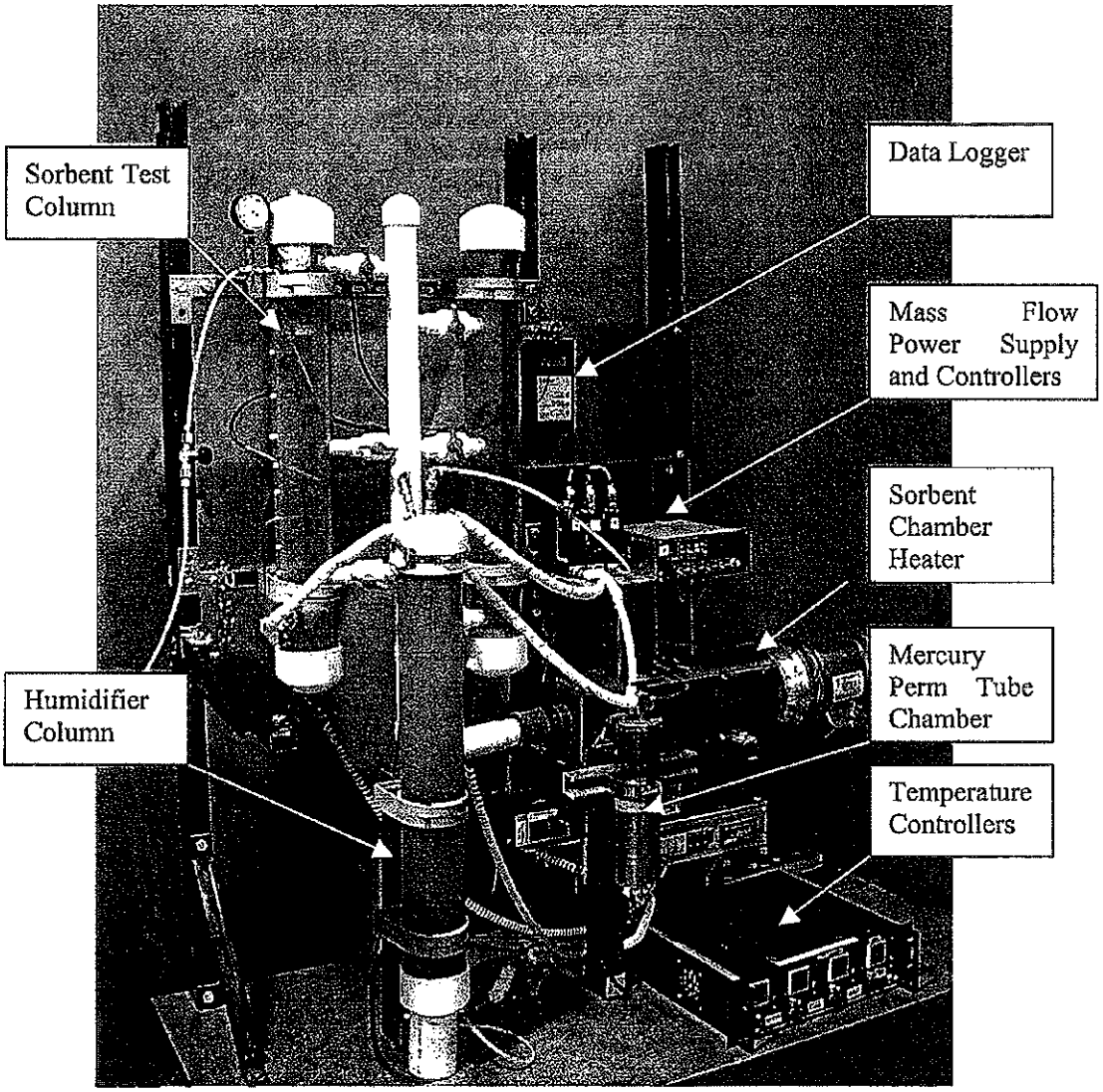

Figure 1. – Carbon Sorption Test Fixture

Sorbent Columns - The sorbent columns were made of 1" diameter clear PVC pipe with a 1-foot active sorbent bed length. The bed diameter was based on industry standard sizing rules, which recommends that the sorbent vessel diameter be roughly eight times the sorbent particle diameter. This ratio of vessel diameter to particle diameter is needed to prevent channeling of test gas through the carbon and to minimize wall effects.

Similar carbon column tests conducted by Nucon were performed in columns of similar size (25mm x 152 mm) (Nucon, 2000). When testing the Calgon Type HGR carbon and the Nucon 3mm MERSORB[7] carbon, 1.5" diameter columns were used to reduce the possibility of channeling.

The sorbent columns were placed inside a 4" diameter clear PVC tube to provide a constant-temperature environment for the sorption tests. Warm air circulated in the annular space to maintain a constant 100°F temperature. Air heated by a finned electric heater was distributed at three points along the length of the 4" pipe. A schematic of the sorption column is shown in Figure 2. The air temperature inside the 4" pipe was monitored and recorded throughout the sorption tests by a data logger.

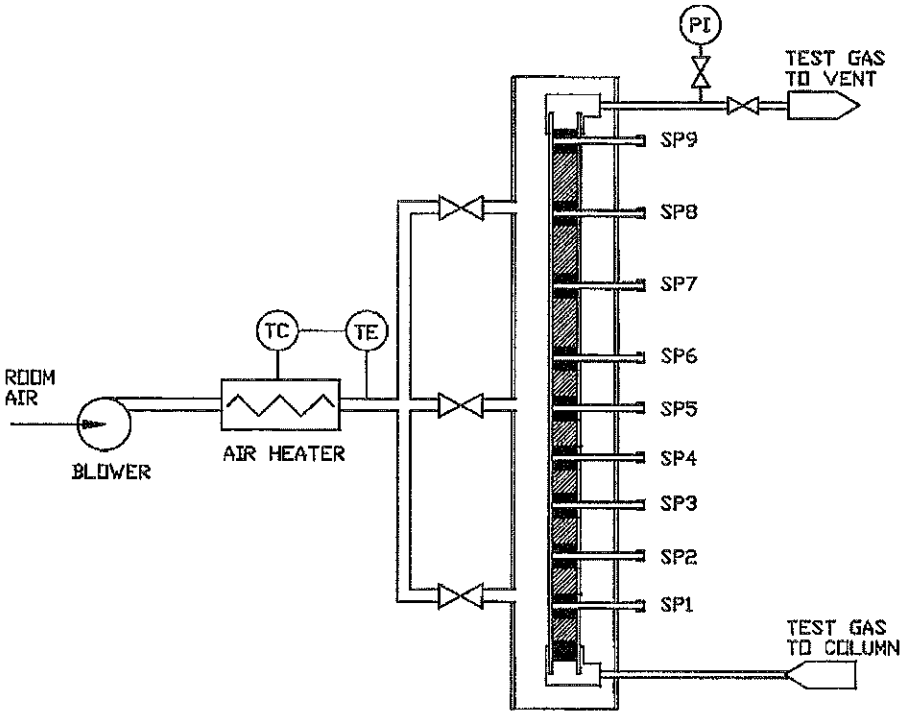

Figure 2. – Schematic of the Sorbent Test Chamber

A series of ¼" sampling ports was installed along the length of the sorbent test chambers so that mercury concentration in the test gas could be monitored as a function of time and bed depth. The presence of the sampling tubes in the carbon bed could alter the flow pattern of test gas in the carbon bed. Therefore, to avoid any gas flow disruptions, the carbon sorption columns were constructed with alternating layers of carbon and non-porous silica glass beads. The glass bead beds provided a zone for inserting the gas sampling tubes. Each of the first six carbon sections were 1" high, followed by a 1" high glass bead zone. Thereafter, the last three carbon beds were 2" high. Sample port were located at 1.5", 3.5", 5.5", 7.5, 9.5", 11.5", 15.5", 19.5", and 23.5" from the gas inlet. A detail of the sampling scheme is shown in Figure 3 below.

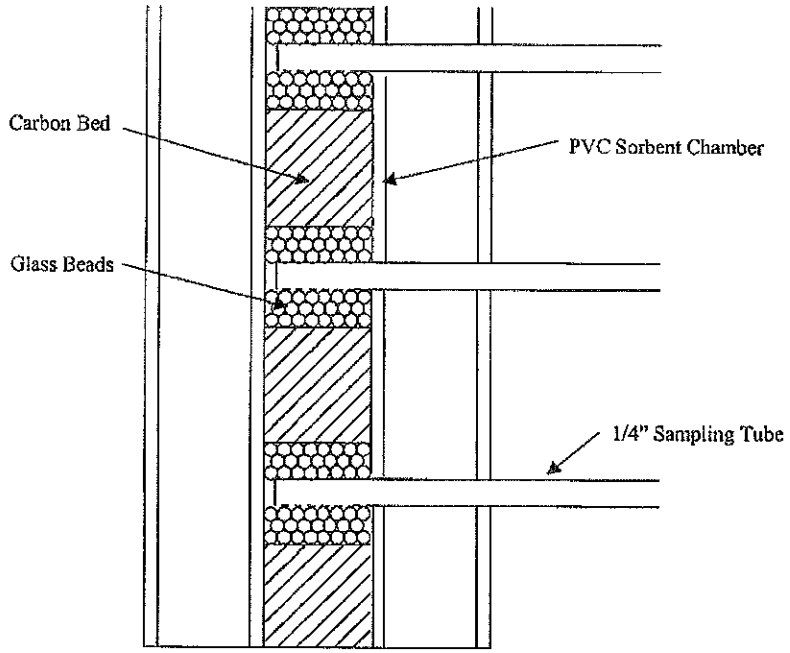

Figure 3. – Carbon Sorption Column

Test Gas Generator - The test gas was generated as shown in Figure 4. Mass flow controllers (MFCs) metered compressed air through a humidification column, mercury permeation tube(s) oven, and a by-pass loop to obtain the desired test gas composition. System pressure for the sorption tests was controlled using a pressure regulator located downstream of the carbon column. A constant pressure of 14.3 psia was maintained during the sorption tests to simulate atmospheric pressure conditions at the SNS facility. The typical atmospheric pressure at the ADA facility in Denver, CO is 12 psia.

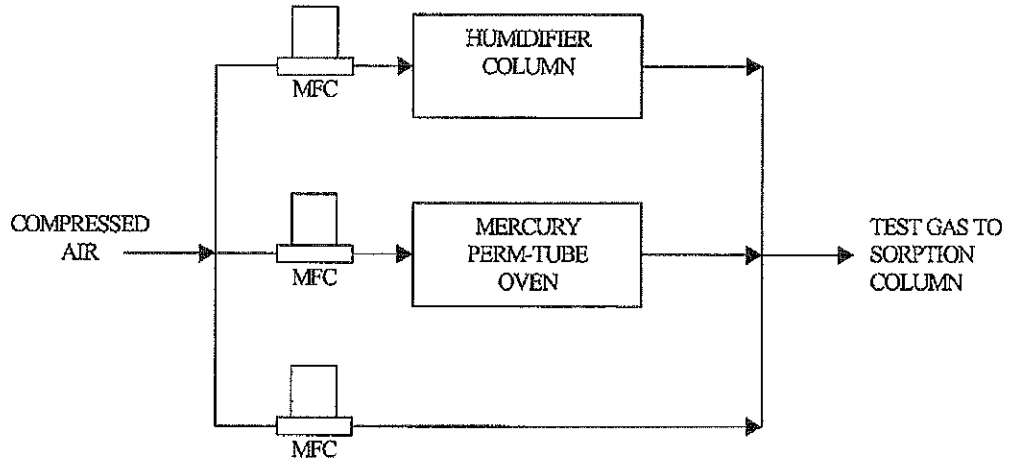

Figure 4. – Flow Schematic for Generating Test Gas

Moisture was introduced by bubbling a portion of the test gas through a humidification column. The humidification column was constructed from a 4" diameter by 3-foot long piece of PVC tube with end caps. An immersion heater was installed in the bottom of the column to heat the water. Temperature of the water was set for each test as a function of the humidity level and gas flow rate. The air exiting the humidifier is nearly saturated with water vapor, so that the final humidity is controlled by blending with a dry air stream in the proper proportion. Humidity level in the test gas was measured by passing the mercury-free air from the humidification column through a container of molecular sieve desiccant. The weight change in the desiccant over the sampling time was used to calculate the amount of water in the air stream. The temperature of the water and flow rate of air through the humidification column was adjusted to achieve the 25% and 50% relative humidity in the test gas.

Mercury permeation tubes provided a source of vapor-phase mercury for the sorption tests. Permeation tubes were purchased through VICI Metronics. The perm tubes are made of ¼" Teflon™ tubing with liquid elemental mercury inside. Caps are used on the ends of the tubing to contain the mercury. Length of the tube varies depending on the desired mercury emission. The mercury source was placed in a larger stainless steel tube and maintained at a constant temperature inside a laboratory oven. A stream of dry gas flowed through the stainless steel tube to transport the mercury-containing gas to the test gas manifold, where it was blended with other gas streams to create the sorption test gas. Adjustments were made as required to maintain a consistent mercury concentration during the sorption tests by changing the temperature of the perm tube oven to achieve the desired 50 $\mu g/m^3$ concentration. The flow rate of nitrogen through the perm tube holder was held constant at 500 sccm.

The emission rate of mercury through the perm tube wall is a function of the tube's geometry and the temperature at which the tube is stored. ADA stores more than 30 perm tubes in temperature-controlled ovens at its laboratory facility. Each month the tubes are weighed to determine the amount of mercury loss. After several months, the average weight loss over time is accurately known (Figure 5). Emission rates are calculated as nanograms of mercury per minute. Target mercury concentration in the feed gas for the sorption tests was 50 $\mu g/m^3$ and was held constant for all carbon sorbent tests.

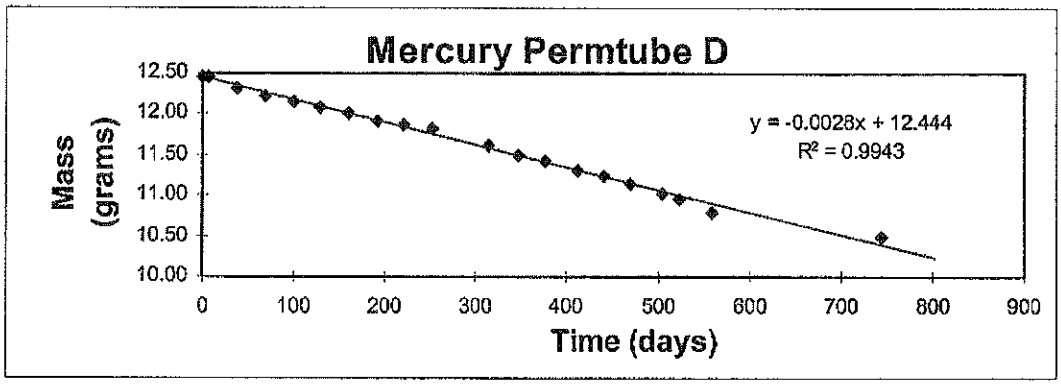

Figure 5. – Perm Tube Calibration Curve

Mercury Analysis for the Sorption Tests

Two methods were used to measure vapor-phase mercury in the test gas. The method used depended on the expected mercury concentration in the gas stream. The first method follows the protocol in U.S. EPA Method 1631, which is used to measure vapor-phase mercury at concentrations less than 0.5 $\mu g/m^3$. In this method, mercury in the sample gas is sorbed onto a gold-coated sand trap. The collected mercury is then released into a stream of argon gas upon heating the gold sand trap. The argon gas is passed through a Tekran Cold Vapor Atomic Fluorescence Spectrometer (CVAFS) which quantifies the amount of mercury in the gas stream. Mercury concentration in the sample gas is calculated by dividing the weight of mercury (nanograms) evolved from the gold sand by the volume of sample gas. This method was used early in the sorption tests to detect the initial mercury breakthrough in the sorbent beds.

The second method used an Ohio-Lumex atomic absorption (AA) instrument to measure vapor-phase mercury. This method is applicable when mercury concentrations are greater than 0.5 $\mu g/m^3$. As in the previous method, mercury from a gas stream is collected onto gold sand and is evolved by heating into a gas stream that feeds the AA instrument. The evolved mercury is passed through an absorption cell illuminated with ultraviolet (UV) light. The attenuation of UV light by mercury is proportional to the amount of elemental mercury in the gas stream. This atomic absorption method was used to measure the inlet mercury concentration to the sorbent test chambers and also to monitor the outlet gas from the sorbent chamber.

The Tekran CVAFS instrument was calibrated using one or more NIST certified mercury solutions. Secondary mercury standards were prepared by diluting one of the stock mercury solutions as prescribed in U.S. EPA Method 1631. Working mercury solutions at different concentrations were then made from the secondary mercury solution and analyzed by the CVAFS instruments. Analysis of the standard solutions established the linear concentration range of each instrument. The Ohio Lumex AA has a vial of elemental mercury built into the instrument that can be rotated into position to measure the mercury vapor concentration in the vapor space. The reported mercury concentration can be checked against values tabulated as a function of temperature.

Gas Sampling Procedure

Test gas from the sorbent columns was extracted through one of several stainless steel sampling tubes inserted above the individual carbon beds. Sample gas was sequentially passed through a gold sand trap to collect the mercury, a mass flow controller to control gas flow through the gold sand trap, and then connected to the house vacuum system to draw gas through the sampling system. Prior to each sampling event, the gas sampling equipment was leak checked by connecting the gold trap and flow meter to the house vacuum system using Teflon tubing. A cap was then placed over the open end of the gold sand trap to draw a vacuum on the sampling equipment. Leakage rate was determined by the mass flow controller reading. Connections and equipment was disassembled and examined if the leakage rate exceeded 10 cc/min.

Once the equipment was leak checked sample gas was flowed through the gold sand trap for a period of 30 minutes. The mass flow controller was set for a gas flow of 300 cc/min. The time at which the flow of test gas was started and ended was record in the project log book. Triplicate traps were taken at a given gas sampling location.

The sorption column has 9 gas sample ports along the length of the column; however, not all of the sample ports were used simultaneously. Primarily two gas sampling ports were used on a weekly basis; the inlet gas port to confirm the inlet mercury concentration and the gas sampling port that corresponded to a 1-second residence time.

Experimental Design

ADA evaluated the mercury uptake capacities for sulfur-impregnated carbons supplied by two carbon manufacturers. Mercury uptake is an important parameter since it indicates how long a carbon material can be in-service at the SNS facility before it needs to be exchanged for fresh carbon. Conditions for the various carbon tests are summarized in Table 2. The effect of gas residence time, gas velocity, carbon manufacturer, carbon particle size, and humidity level of the test gas on the mercury capacities were examined on this project.

During the sorption tests, relevant operational data for the experiment was monitored on a daily basis including the average sorbent chamber temperature and pressure in the sorbent column. Inlet mercury concentration was monitored with the Ohio Lumex AA on nearly a weekly basis. Adjustments were made as required to maintain a consistent mercury concentration during the sorption tests. Mercury measurements at appropriate sample ports were made on a nearly weekly basis to track the mercury mass transfer zone through the carbon columns. As mentioned previously, CVAFS and AA methods were used to measure the amount of mercury in the test gas.

Mercury breakthrough curves for the carbon tests were prepared using the experimental data and a computerized version of the Rosen fixed-bed sorption model. This model was developed in the 1950s by J. B. Rosen (Rosen, 1952) and takes into account both the rate mechanisms to transfer adsorbate (i.e., mercury) between the gas and solid phases and the rate mechanism involving the transfer of material within a spherical solid phase sorbent particle. The computational form of the Rosen model used for the analysis was taken from Ruthven (Ruthven, 1984). The model parameter of major importance is K_d which represents the capacity of the sorbent material for a given adsorbate. The Rosen model uses a linear adsorption isotherm to predict the partitioning of mercury between gas and solid phases. The mathematical form of this type of isotherm is as follows:

$$q = K_d (C_{Hg})$$

where, q is the equilibrium mercury loading on the solid phase or sorbent expressed as weight of mercury per weight of sorbent, K_d is the proportionality constant with units of volume of gas treated per weight of sorbent, and C_{Hg} is the equilibrium mercury concentration in the gas phase. The magnitude of K_d represents the mercury capacity of the sorbent material.

The Rosen model was also used to predict the time for the outlet gas of a full-scale mercury filter system to reach a "critical" breakthrough mercury concentration of 0.1

Table 2. – SNS Sorbent Test Matrix

		Test 2	Test 3	Test 4	Test 5	Test 6
Carbon Type		Nucon 1.5mm	Nucon 1.5mm	Nucon 3mm	Nucon 1.5mm	Calgon HGR
Input Hg Concentration (ug/m3)		50	50	50	50	50
Relative Humidity %		25	25	25	50	25
Gas Flow (L/m)		4.7	2.3	10.5	4.7	10.5
Velocity (Ft/Min)		30	15	30	30	30
Sample Port	Carbon Bed Height (in)	Calculated Residence Times in Seconds				
SP1	1	0.2	0.3	0.2	0.2	0.2
SP2	1	0.3	0.7	0.3	0.3	0.3
SP3	1	0.5	1.0	0.5	0.5	0.5
SP4	1	0.7	1.3	0.7	0.7	0.7
SP5	1	0.8	1.7	0.8	0.8	0.8
SP6	1	1.0	2.0	1.0	1.0	1.0
SP7	2	1.3	2.7	1.3	1.3	1.3
SP8	2	1.7	3.3	1.7	1.7	1.7
SP9	2	2.0	4.0	2.0	2.0	2.0
Column Diameter (in)		1.0	1.0	1.5	1.0	1.5

$\mu g/m^3$. The full-scale system will be designed to operate at a gas velocity of 15 feet per minute. However, most of the carbon tests were conducted at a gas velocity of 30 feet per minute. Using the Bed Depth Service Time approach, it was possible to take the experimental test results conducted at one gas velocity and predict what will happen at a different gas velocity. The Bed Depth Service Time model uses the irreversible isotherm model of Bohart and Adams to correlate breakthrough times and breakthrough concentrations as a function of adsorption capacity, inlet mercury concentration and gas velocity (Cooney, 1999). For our purposes, the Bed Depth Service Time (BDST) approach was used to translate tests results conducted at a gas velocity of 30 feet per minute to estimate the lifetime of carbon in a full-scale mercury filter system operating at a gas velocity of 15 feet per minute.

The BDST model takes the mathematical form:

$$t_b = \frac{N_o}{\varepsilon \upsilon C_o} D - \frac{\ln\left(\frac{C_o}{C} - 1\right)}{kC_o}$$

where t_b is the breakthrough time, N_o is the adsorption capacity of the adsorbent per unit volume of bed, ε is the porosity of the bed, υ is the interstial gas velocity, C_o and C are the inlet and outlet mercury concentrations, respectively, D is the sorbent bed depth, and k is mass transfer rate constant. This equation shows that the breakthrough time is a linear function of the bed depth, with $\frac{N_o}{\varepsilon \upsilon C_o}$ as the slope of the line and $\frac{\ln\left(\frac{C_o}{C} - 1\right)}{kC_o}$ as the intercept. The equation can be used to predict breakthrough times for changes in gas flow rate through the sorption bed. The value of the rate constant k is determined by the net effect of mass transfer of an adsorbate from the gas stream to a sorbent particle and by solid-phase mass transfer of the adsorbate on the surface of the sorbent particle. Fluid-phase transport is characterized by the fluid mass transfer coefficient, which typically varies with fluid velocity to the 0.6 power. Solid-phase mass transfer on and in a sorbent particle is characterized by the surface diffusivity coefficient and is independent of gas velocity. Assuming the solid-phase mass transfer resistance dominates, then k is not significantly affected by changes in gas velocity (υ), and thus the intercept in the BDST model remains essentially unchanged when gas velocity is changed. However, the slope in the BDST equation certainly does change with υ. The effect of changes to gas velocity on breakthrough time can be predicted by calculating a new value for the slope using the following equation.

$$New\_Slope = Old\_Slope\left(\frac{\upsilon_{old}}{\upsilon_{new}}\right)$$

Discussion of Test Results

The five carbon sorption tests were run as specified in the test matrix. Inlet mercury concentration for these tests is plotted as a function of days of service in Figure 6. The average inlet mercury concentration was 55.4 $\mu g/m^3$ with a 4.5 $\mu g/m^3$ standard deviation. Target mercury concentration for these tests was 50 $\mu g/m^3$. Average inlet mercury concentrations for the individual tests are shown as bold lines through the data groupings. Variation in mercury concentration is a result of temperature changes made to the perm tube chamber to accommodate different feed gas flow rates for the sorption tests.

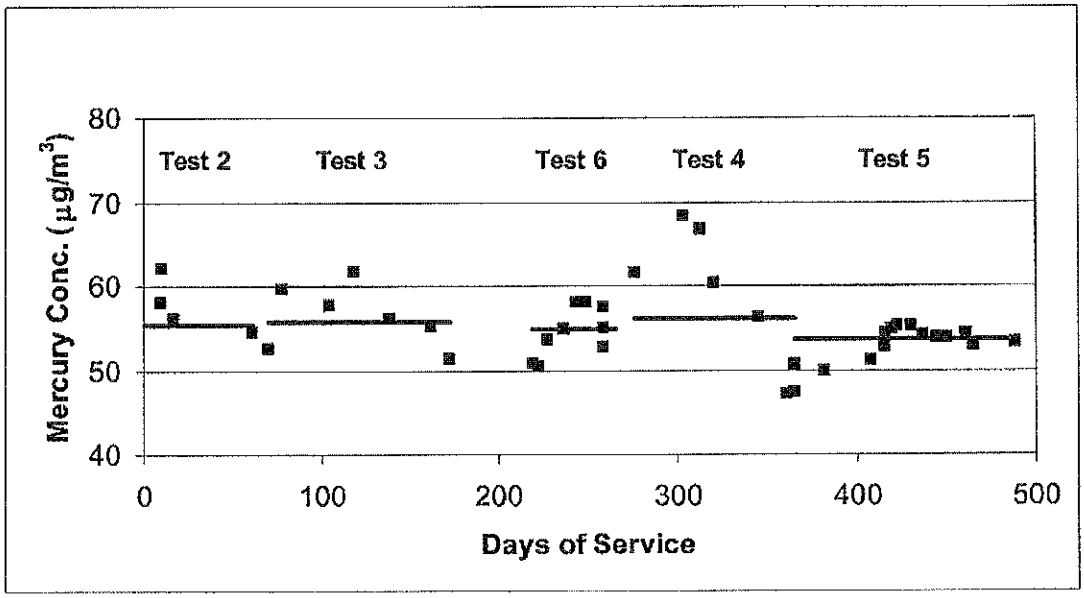

Figure 6. – Inlet Mercury Concentration for Carbon Tests

The duration of each test varied depending on the carbon performance. Tests were terminated when the mercury concentration at a 1-second residence time reached a value of 0.1 $\mu g/m^3$. For tests conducted at superficial gas velocities of 15 and 30 feet per minute, the 1-second residence time corresponded to a carbon height of 3 and 6 inches, respectively. The mercury data from each test were used to estimate the mercury sorption capacity of the carbon sorbent and to model the theoretical breakthrough curves. Breakthrough curves were generated using the Rosen model. Breakthrough curves were predicted for each carbon sorbent by varying model parameters such that the resulting curves were representative of the experimental mercury breakthrough data. Figure 7 shows the predicted breakthrough curves for 1", 2", 3", and 4" carbon beds in Test 2.

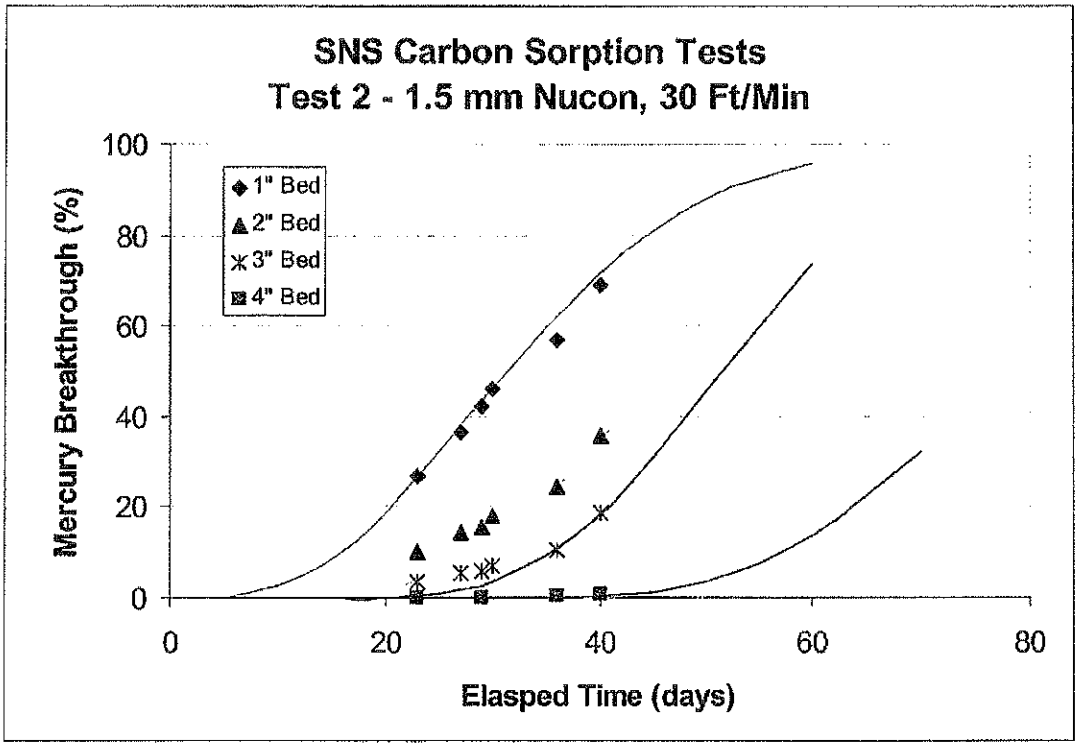

Figure 7. –Breakthrough Curves Predicted with the Rosen Model

Table 3 summarizes the model parameters used and results calculated from the model for each carbon test. The objective of these carbon tests was to evaluate changes in mercury capacity as a function of carbon manufacturer, gas flow rate, particle size of the sorbent, and humidity in the test gas. Each comparison is discussed in turn below.

Comparison of Carbon Manufacturers

Carbon samples used for this comparison were the Nucon 1.5mm MERSORB[7] carbon and the Type HGR carbon from Calgon (Figure 8). The carbon samples were not very similar in a physically sense since the Nucon 1.5mm carbon was a cylindrical pellet and the Type HGR Calgon carbon was an irregularly shaped granular material. The diameter for an equivalent spherical particle was calculated for each carbon by determining the volume of an individual carbon particle. The calculated particle sizes for the Nucon and Calgon carbons based on a spherical shape were comparable at 0.09 and 0.11 inch, respectively. Both Test #2 (Nucon) and Test #6 (Calgon) were conducted at a superficial gas velocity of 30 feet per minute and test gas with 25% relative humidity.

The K_D values calculated at these test conditions for the Nucon 1.5mm MERSORB[7] and Calgon Type HGR carbon were 23 and 17 million liters per kilogram of sorbent, respectively. The mercury capacity for the Nucon carbon was slightly higher than the Calgon carbon and is reflected in the estimated lifetime for carbon in a full-scale system. At an operating superficial flow rate of 15 feet per minute, the Nucon 1.5mm MERSORB[7] carbon would maintain mercury concentrations below 0.1 $\mu g/m^3$ for 400

Table 3. – Summary of SNS Carbon Test Results

Test No.	Carbon Type	Measured Diameter (inch)	Porosity (%)	Tortuosity	Particle Density (g/ml)	Superficial Velocity (ft/min)	Relative Humidity (%)	Mercury Capacity (l/kg)	Lifetime at 15 ft/min (days)
2	Nucon Mersorb 1.5mm	0.09	60	0.2	0.85	30	25	23 MM	400
3	Nucon Mersorb 1.5mm	0.09	60	0.2	0.85	15	25	34 MM	540
4	Nucon Mersorb 3mm	0.16	60	0.2	0.92	30	25	25 MM	320
5	Nucon Mersorb 1.5mm	0.09	60	0.2	0.85	30	50	52 MM	900
6	Calgon HGR-granular	0.11	60	0.1	1.15	30	25	17 MM	260

days, whereas for the Calgon Type HGR carbon, mercury concentrations are expected to be greater than 0.1 $\mu g/m^3$ after 260 days.

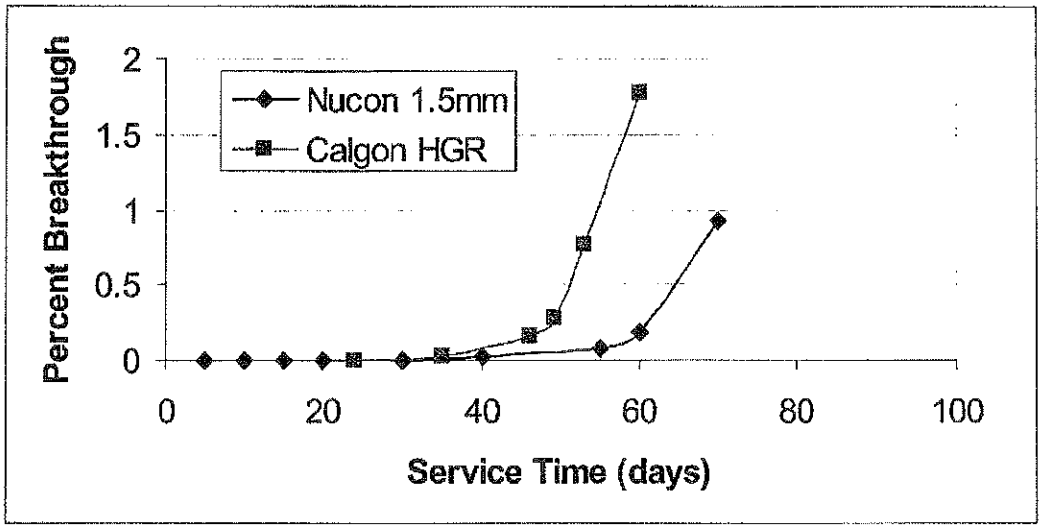

Figure 8. – Performance of Nucon 1.5mm and Calgon HGR Carbons

Effect of Gas Flow Rate on Mercury Capacity

Effect of gas flow rate on the sorption capacity was studied in Test #2 and Test #3 using the Nucon 1.5mm MERSORB[7] (Figure 9). Gas rates used in these tests were 15 and 30 feet per minute. Relative humidity in the test gas was the same at 25% RH.

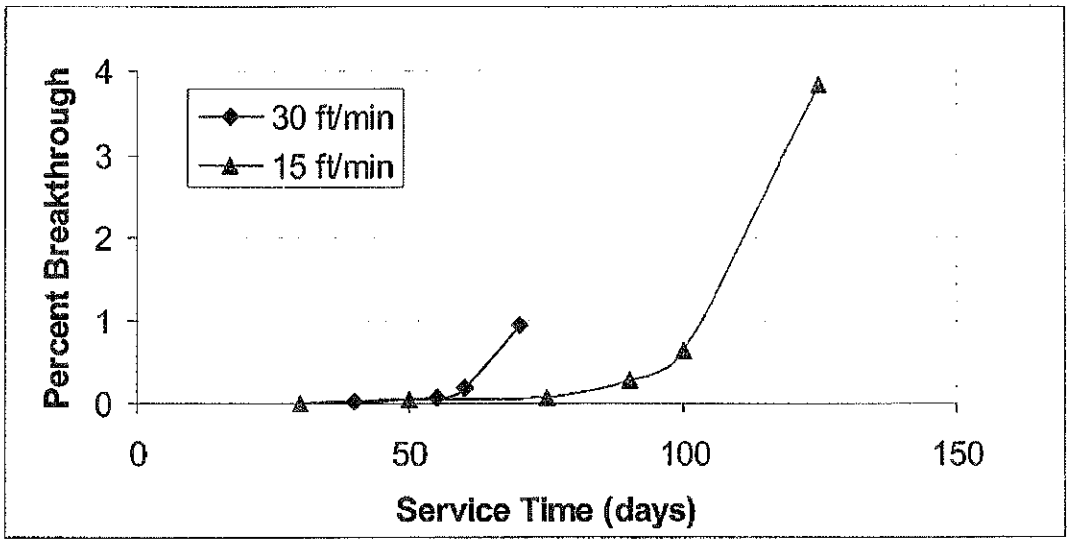

Figure 9. – Performance of Nucon 1.5mm at Different Gas Flow Rates

The K_D values calculated for Test #2 and Test #3 were 23 and 34 million liters per kilogram of sorbent, respectively. The Nucon carbon used in a system operating at the lower flow rate of 15 feet per minute had nearly 50% more mercury capacity compared to

a system operating at the higher flow rate of 30 feet per minute. In a full-scale system operating at a superficial flow rate of 15 feet per minute, the Nucon 1.5mm MERSORB[7] carbon should maintain mercury concentrations below 0.1 $\mu g/m^3$ for 540 days compared to 400 days for a mercury capture filter system operating at superficial gas velocity of 30 feet per minute.

This result is reasonable given that as fluid velocity increases, the adsorbate concentration profile (mass transfer zone) in the carbon bed becomes broader and a greater amount of the sorbent is not well utilized. In systems operating at higher fluid velocities, breakthrough will occur earlier than anticipated and carbon sorbent will need to be changed out sooner than for a system operating at a lower fluid velocity.

Effect of Particle Size on Mercury Capacity

Effect of particle size on the sorption capacity was studied in Test #2 and Test #4 using the Nucon 1.5mm and Nucon 3mm MERSORB[7], respectively (Figure 10). Gas rates used in these tests were the same at 30 feet per minute. Relative humidity in the test gas was also the same at 25% RH.

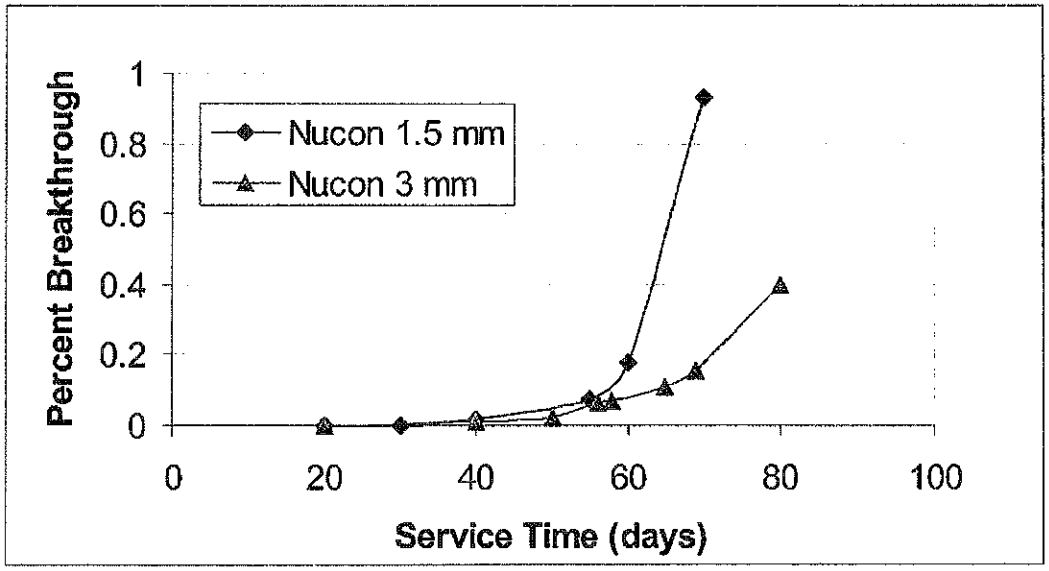

Figure 10. – Performance of Nucon 1.5mm and 3mm Carbons

The K_D values calculated for Test #2 and Test #4 were 23 and 25 million liters per kilogram of sorbent, respectively. The mercury capacities for the two Nucon carbons were nearly the same. The actual height of carbon in the columns for tests #2 and #4 corresponded to 0.91 and 1.22 seconds residence time, respectively. Normalizing the results for a 1 second residence time, the 1.5mm carbon had better performance compared to the 3mm carbon. For a full-scale system operating at a superficial flow rate of 15 feet per minute, the Nucon 1.5mm MERSORB[7] carbon should maintain mercury concentrations below 0.1 $\mu g/m^3$ for 400 days compared to 320 days for a mercury filter system using the 3mm carbon.

The results from these tests are inline with those published by Nucon International (Nucon, 2000). In their report, Nucon found that an increase in particle size negatively

affected the performance of their carbons. Tests were performed with three different sizes of carbons; 1.5mm, 3mm, and 4mm and all carbon columns were operated at a superficial velocity of 6 feet per minute. The initial mercury capture efficiency was measured at a bed position equivalent to a 1.67 seconds residence time. The initial removal efficiencies for the three carbons were 100%, 92% and 76%, respectively.

Effect of Humidity on Mercury Capacity

Effect of humidity in the test gas on the sorption capacity was studied in Test #2 and Test #5 using the Nucon 1.5mm MERSORB[7] carbon (Figure 11). Gas rates used in these tests were the same at 30 feet per minute. Relative humidity in the test gas for Test #2 was 25%, and 50% for Test #5.

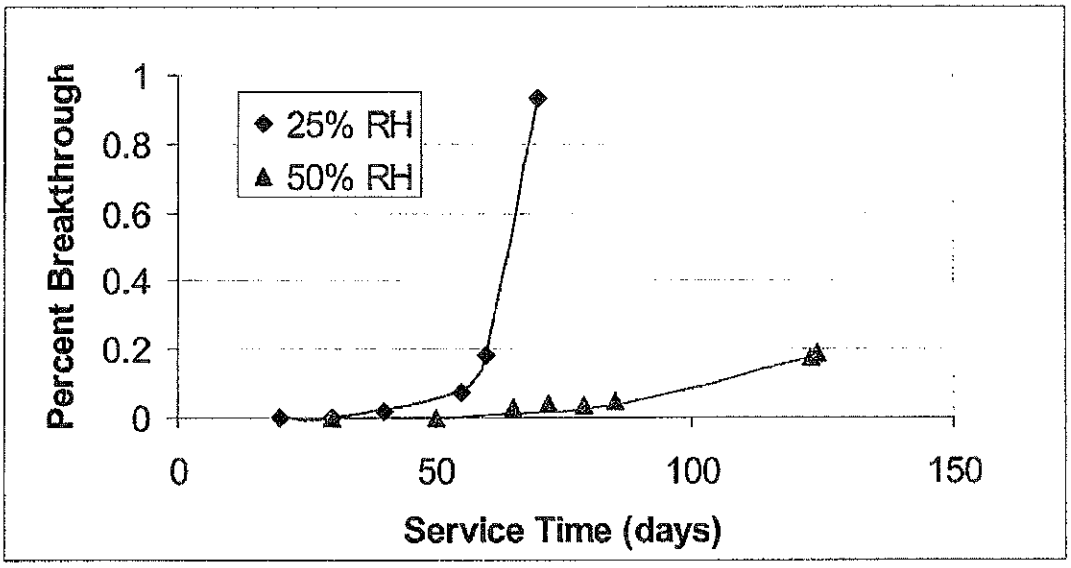

Figure 11. – Performance of 1.5mm at Different Relative Humidities

The K_D values calculated for Test #2 and Test #5 were 23 and 52 million liters per kilogram of sorbent, respectively. The mercury capacity for Test #5 was substantially higher compared to that determined for the 1.5mm carbon in Test #2 and is reflected in the estimated lifetime for carbon in a full-scale system. For a full-scale system operating at a superficial flow rate of 15 feet per minute and with 50% RH, the Nucon 1.5mm MERSORB[7] carbon should maintain mercury concentrations below 0.1 $\mu g/m^3$ for nearly 900 days compared to a lifetime of 400 days for a mercury filter system using the 1.5mm carbon at 25% RH.

Summary and Conclusions

ADA tested three carbon sorbent materials from two carbon manufacturers in controlled laboratory tests. The two carbon manufacturers were Nucon International and Calgon Carbon Corporation. The main operating parameters studied in the mercury sorption tests were superficial gas velocity, sorbent particle size, and humidity in the test

gas. The purpose of these tests was to determine the mercury capacities of the carbon sorbents under various operating conditions. Results of the carbon tests are summarized below.

- The Nucon 1.5mm carbon had superior mercury capacity compared to the Calgon Type HGR material.
- Carbon column operating at a superficial velocity of 15 feet per minute had better performance compared to a column operating at a superficial velocity of 30 feet per minute.
- Carbon sorbent with a smaller particle size (1.5mm) had better performance than a carbon sorbent of larger particle size (3mm).
- Carbon column operating at a relative humidity of 50% outperformed a column operating at a relative humidity of 25%.

References

NUCON Bulletin 11B28, "Design and Performance Characteristics of MERSORB[7] Mercury Adsorbents," January 2000.

Cooney, David, O., "Adsorption Design for Wastewater Treatment," CRC Press LLC, 2000 Corporate Blvd., N.W., Boca Raton, Florida, 33431, p 162, 1999.

Calgon Carbon Corporation Type HGR Carbon Product Bulletin LC-27-204a, 1997.

McCabe, W. L., J. C. Smith, P. Harriott, "Unit Operations of Chemical Engineering," McGraw-Hill chemical engineering series, McGraw-Hill, Inc., New York, NY, 1985.

Ruthven, D. M., Principles of Adsorption and Adsorption Processes, Wiley & Sons, New York, NY, 1984.

Rosen, J. B., Kinetics of a fixed bed system for solid diffusion into spherical particles, Journal of Chemical Physics, v20, n387, 1952.