Final Report

ARB Contract: # 95-348

PROTOTYPE DEMONSTRATION OF CHA NOx REMOVAL SYSTEM FOR TREATMENT OF STATIONARY DIESEL ENGINE EXHAUST

Prepared for:
California Air Resources Board
and the **California Environmental Protection Agency**

Prepared by:
CHA Corporation
Laramie, Wyoming 82070
Principal Investigator: Dr. Chang-Yul Cha

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Disclaimer

Acknowledgments

We wish to express our appreciation to the California Air Resources Board for supporting this project. Special thanks goes to Marla Mueller for her guidance in organizing the project. We are also grateful for the input and support received from Rick Wiesner of SMUD, Phil Mook and John Carroz of the U. S. Air Force, McClellan AFB, and John Gerling with Gerling Applied Engineering.

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ABSTRACT

CHA Corporation has designed, fabricated, developed and demonstrated an integrated prototype system capable of treating diesel engine exhaust for a 58 hp diesel engine. The integrated system is a continuous adsorption/ regeneration system capable of 100% NOx removal, as well as destroying the VOCs and soot present in diesel exhaust. The pollutants are first removed from the flue gases by passing the gases through a bed of relatively inexpensive carbon adsorbent. The NOx, SOx, and VOC gases are adsorbed and stored on the carbon during this step. The pollutants are then destroyed during regeneration of the carbon by using microwave energy. The microwave energy decomposes NOx to nitrogen and carbon dioxide, VOCs to carbon dioxide and water, and SOx to elemental sulfur and carbon dioxide. A prototype unit was constructed and perfected through thorough experimentation. The unit was then transported to McClellan Air Force Base in Sacramento, CA in March of 1998 for a week long demonstration. The field demonstration of the prototype was very successful. No mechanical or technical problems were encountered with frequent start up and shut down operations. During the week of field demonstration, the outlet gas did not contain any measurable NOx.

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1.0 Introduction

The CHA Corporation has developed a new process for removing and destroying pollutants from combustion flue gases. The primary pollutants of interest are nitrogen oxides (NOx) and sulfur oxides (SOx) which are commonly present in combustion flue gases[1] and are strictly regulated by the Clean Air Act. The overall objective of this project was to design, build and demonstrate a prototype devise utilizing the CHA process to remove NOx and other pollutants from the exhaust gases emitted by a 58-hp diesel engine.

Currently, there is no single device available for the simultaneous removal of NOx, CO, VOCs and PM10 from oxygen-rich exhaust gases. Two methods for NOx reduction are currently available: selective non-catalytic reduction (SNCR) and selective catalytic reduction (SCR)[2]. In SNCR and SCR (processes originally developed for large, coal-fired power plants), a chemical reducing agent, usually anhydrous ammonia or urea, is injected into the exhaust gas stream as it exits the combustor. In the SCR process, impregnated gases pass through a catalytic element that promotes reaction of NOx with the reducing agent to form nitrogen and water. The SCNR process has a much lower NOx-removal efficiency and requires higher temperatures than SCR. SCR vendors report that a 90% reduction in NOx levels is achievable if the catalyst bed can be operated at temperatures between 400 and 500 °C. However, the costs of implementing and operating the SCR process are very high, and emissions of small amounts of ammonia are very likely. Currently, a low-temperature (ambient to 125 °C) NOx-removal technology is not available. The CHA process is intended to fill this need.

The emission of CO and VOCs can be reduced by a catalytic converter very similar to that found in automobiles[3]. A catalytic converter is a ceramic honeycomb structure that is impregnated with oxidation catalysts, usually platinum and/ or palladium. When a hot exhaust stream passes through the ceramic structure, the catalyst promotes the reaction of CO and VOCs with oxygen in the exhaust gases. Catalytic converters are most effective when the catalyst element is above 260 °C with optimal performance near 400 °C[4,5,6,7]. We assume PM10 is mainly carbonous material that is produced by internal combustion engines, especially diesel engines. The soot can be either oxidized or removed by a filter. In catalytic oxidation, the exhaust gas is sent through a catalyst element, which traps and oxides the soot. Removal efficiencies of up to 50% are achievable with this process the average exhaust temperature can be kept near 400 °C[8]. Generally, higher removal efficiencies are not possible due to the temperature limitations of the exhaust stream. Very high removal efficiencies are achievable with soot filters. These paper filters for low-temperature service or ceramic fiber filters for high-temperature service remove soot from the exhaust stream as it flows through the filter. Typical service lifetimes for these filters are short, usually less than 50 hours. The process developed and reported here fills the need for a new process to deal with the harmful emissions from diesel engines.

Dr. C.Y. Cha first discovered the CHA process in the late 1980's. Follow-up exploratory work was performed under the sponsorship of the United States Department of Energy (DOE) under Phase I and Phase II SBIR grants (DOE contract number DE-FG03-90ER80898) and was completed in August of 1993. The process is disclosed in four US patents (numbers 5,246,554; 5,256,265; 5,269,892, process; 5,269,892, equipment)[9,10,11,12].

The process can be utilized on gas streams that contain NOx, SOx, or both NOx and SOx. The pollutants are first removed from the flue gases by passing the gases through a bed of relatively inexpensive carbon based adsorption catalyst. The NOx and SOx gases are adsorbed and stored on the adsorption catalyst during this step. The pollutants are then destroyed during regeneration of the catalyst with microwave energy. The microwave energy decomposes NOx to nitrogen and carbon dioxide and decomposes SOx to elemental sulfur and carbon dioxide[13].

Laboratory results indicate that this catalyst shifts the equilibrium of the exhaust gases so that species that are not normally well adsorbed by the high surface area carbon adsorbent, such as NOx, are readily adsorbed while the carbon's natural affinity for substances such as SOx, soot, and VOC's is left intact[14].

Regeneration can be conducted either in place or in a separate vessel, depending upon the overall system requirements. Microwave energy is introduced during regeneration through a simple system that is conceptually identical to a home microwave oven. This energy is readily absorbed by the pollutant-laden adsorbent and promotes NOx and SOx reactions with carbon to decompose NOx into nitrogen and carbon dioxide and SOx into elemental sulfur and carbon dioxide.

Because the energy transfer is through electromagnetic means rather than bulk conventional heating, we have consistently observed that the bulk gas temperature remains remarkably low throughout regeneration, usually less than 300 °F (149 °C). Once regeneration is completed, the adsorbent is ready for reuse. Total removal efficiencies of 98% and greater have been experimentally observed for NOx removal, SOx removal, and combined NOx and SOx removal[15].

2.0 Materials and Methods

The design for a working prototype unit was based on the emissions of a 58 hp diesel engine. The conceptual design, however, is applicable to larger applications. The design criterion put forth by CHA Corporation and partners, SMUD, McClellan Air Force Base, and Gerling Engineering, to control emissions from diesel power carts is as follows:

1. The existing power carts can not be modified in any way.
2. The NOx abatement unit should be a stand alone, self powered unit on wheels that is no larger than the existing power carts.
3. The connections of the exhaust piping from the power cart to the device should be through a simple flexible hose or other means that can be easily disconnected and left behind if the power cart(s) need to be mobilized elsewhere.
4. Power is not always available where the power carts are used. If a light cart is used in a given ground operation effort, then 10 amps of 120 volt 60 cycle power is available.
5. The device should be designed to operate six months without regular maintenance.

Design criterion was set forth for standby generator applications as follows:

1. Emissions from the unit should not exceed 1.5×10^{-4} pounds NOx/hours/kW.
2. Post treatment emissions from a 250 kW unit not to exceed 0.038 pounds NOx/hour, or 0.076 pounds NOx/hour from a 500 kW unit.
3. NOx removal efficiency to be at least 90 percent.
4. The only operation of the unit will be intermittent such as stand-by power generators. The required demonstration, maintenance, emergency and Auxiliary Power Program generators are typical applications. Total operation less than or equal to 175 hours per year.
5. Design lifetime should be 30 years.
6. A 250 kW rated unit must be devised which will operate for 30 years at a present value cost of $162,900.
7. Operation shall be unattended, except for periodic servicing.
8. Devise must be able to satisfy regulators that emissions will not exceed guarantees and/ or permit limits.
9. To minimize cost and operating complexity, the installation should not require continuous emission monitoring equipment.

10. Device should fit in existing exhaust piping without exceeding engine manufactures back pressure limits.
11. Device should be suitable for outside installation and servicing at either ground or rooftop levels.
12. Exhaust cooling features should be passive and rated to function in no-wind conditions at up to 105 degrees Fahrenheit.
13. Device should be Underwriters Laboratories rated for fire, and electrical and microwave safety.

The main objective of the project was to incorporate the CHA NOx Removal Process in a system that would remove and destroy NOx, VOC, and soot pollutants from the exhaust of a 58 hp diesel engine. Diesel exhaust gas is composed of the following[1]:

Table 1 : Diesel Exhaust Gas Composition

Component	Amount
Carbon Monoxide (CO)	736 ppm
Nitric Oxide, NO	1287 ppm
VOCs	303 g/h
Soot	1.32 g/hp.h
H_2O	7.20%
Carbon Dioxide, CO_2	7.38%
Oxygen, O_2	9.06%
Nitrogen, N_2	76.10%

The main goal of this study was to design and develop an integrated prototype to remove NOx from diesel engine exhaust gas. In order to meet this goal, the development work was broken down into the following nine tasks:

Task 1: Prototype Design
Task 2: Fabrication of Prototype Device
Task 3: Installation and Develop Test Plan
Task 4: Start-up and Shakedown Operations
Task 5: Test Soot Destruction Device
Task 6: Fabricate Regeneration Device and Build Heat Exchanger
Task 7: Test Heat Exchanger and NOx Adsorption
Task 8: Study Gas Residence Time and Particle Size
Task 9: Demonstration, Technical and Economic Evaluation and Final Report

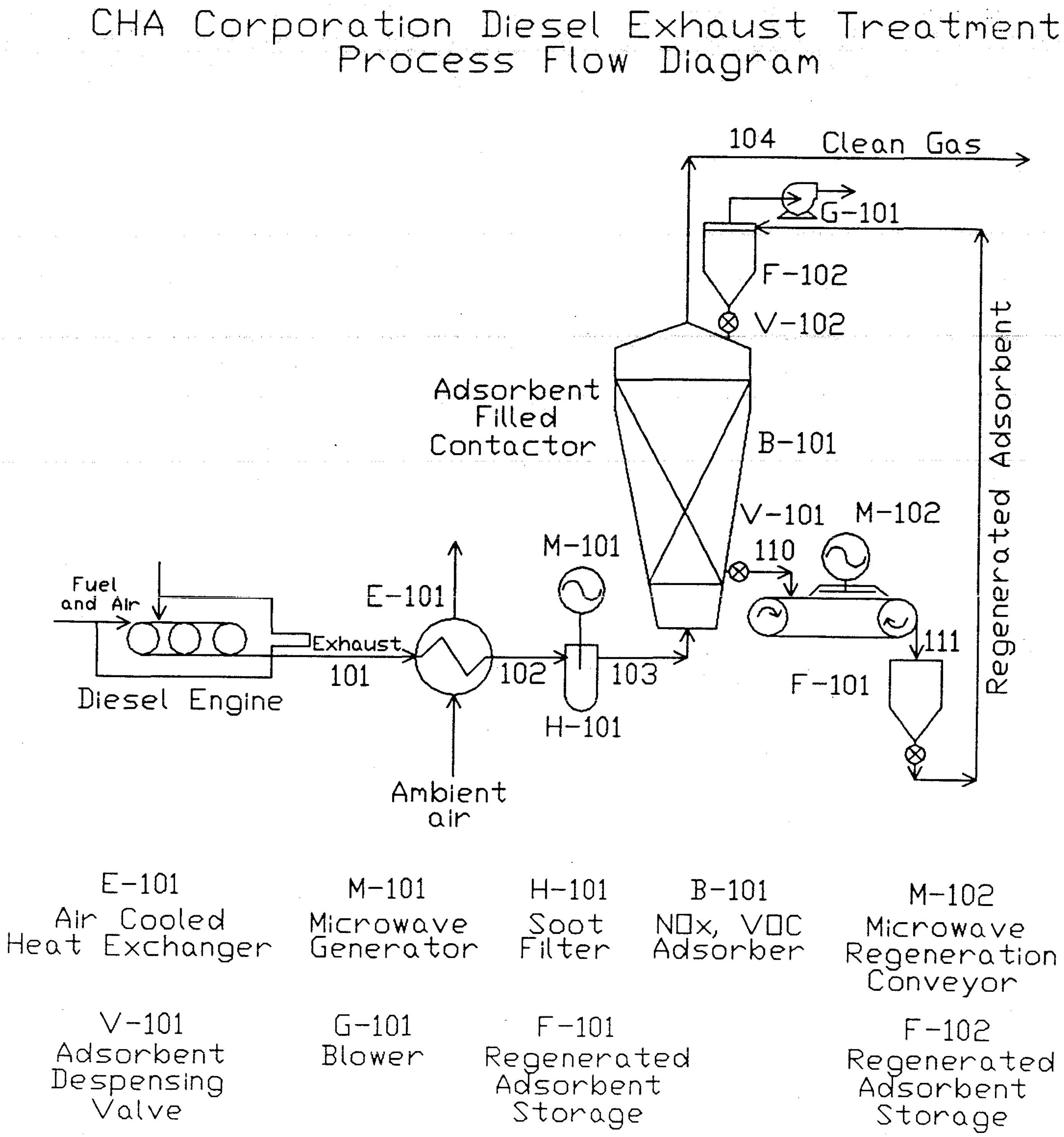
Task 9 addresses the technical and economic evaluation and details from that task are presented in the Results and Discussions section. The results obtained from all other tasks are presented below.

2.1 Prototype Design, Construction, Installation, and Shakedown

Tasks 1-4 focused on the design, development and installation of a prototype design capable of removing NOx from diesel engine exhaust gas. The purpose of this prototype device was to incorporate the CHA NOx removal process in a system that removes and destroys NOx, VOCs, and soot pollutants from the exhaust gas of a 58 horsepower engine. As a part of Task 1, the service and engineering specifications for a conceptual commercial unit were defined. Figure 1 shows the first process flow diagram designed to accommodate a 58 hp diesel engine. The engine is coupled to a 35 kW three phase 240 volt generator. The engine is loaded by applying generated electrical current to a resistive load bank. The engine is a naturally aspirated, four cylinder, four cycle water cooled unit manufactured by John Deere Company. This engine produces 110 SCFM (187 sm^3/h) of exhaust gas at temperatures ranging from 350 degrees to 1080 °F (177-582 °C) depending on engine load. NOx production is from 150 ppm at zero load to 2600 ppm at 100% load. Soot production is 5.6 grams per hour at 50% load. Volatile organic compound (VOC) production is 90 grams per hour at 50% load. The exhaust treatment device is sized for this engine but can be easily fitted to other similar sized diesel engines.

Two types of batch adsorbers were intially evaluated. The first batch adsorber was a tapered vertical cylinder five feet (1.5 m) tall and 35 inches (0.9 m) in diameter at the large end and 18 inches (5.5 m) in diameter in the small end. The taper discourages any wall effects that allow the exhaust gas to bypass the adsorbent. The adsorber was custom made and holds 400 pounds of char adsorbent. A stainless steel support screen holds the carbon 12 inches (30.5 cm) above the bottom of the adsorber. Gas was routed from the bottom to the top. The second batch adsorber design is a radial flow scheme. The radial adsorber considered is 15 inches (38.1 cm) in diameter and 48 inches (1.22 m) tall. Gas flows from outside, the larger diameter, to inside. The outside housing of the radial adsorber is equipped with one-inch (2.5 cm) steel fins that help cool the exhaust gas.

Figure 1: Prototype Design for Treatment of Diesel Engine Exhaust



		101	102	103	104	110	111
Phase		Vapor	Vapor	Vapor	Vapor	Solid	Solid
Temperature	F	650	175	170	160	170	200
Pressure	In. H2O	15	13	8	0	0	0
Flowrate	SCFM	100	100	100	100		
	Lb/Hr					50	50

After extensive testing of the batch adsorber, it was concluded that the adsorption capacity of activated carbon or char is too small for the batch adsorber to be able to last for a reasonable amount of time. As a result, the batch adsorber (either radial or vertical) was changed to the continuous moving bed reactor.

The moving bed adsorber is 30-inches x 30-inches x 58-inches (76.2 cm x 76.2 cm x 147.3 cm) tall, square with rounded corners. The gas is introduced through a 2-inch (5 cm) diameter distribution manifold pipe, located 20-inches (51 cm) diameter distribution manifold pipe, located 20-inches (51 cm) above the base. The details of the moving-bed adsorber will be described in the integrated prototype system.

Removal of the NOx and VOC pollutants is accomplished by passing the exhaust gas through a bed of carbon based adsorbent bed where the pollutants are selectively adsorbed and stored. Removal of the soot is accomplished by passing the exhaust gas through a dedicated soot filter. Once collected, the NOx and VOC pollutants are exposed to microwave energy by passing the carbon based adsorbent through a microwave energy field where the pollutants are decomposed. The decomposition products are carbon dioxide, nitrogen and water. Fresh or regenerated adsorbent is added to the contactor each time adsorbent is removed. The adsorbent is re-used after this exposure to microwaves (regeneration). The soot captured by the filter is periodically converted to carbon dioxide by introducing microwave energy to the filter. Microwaves selectively heat the captured soot and accelerate oxidation of the soot without directly heating the filter material.

As a part of Task 2, a prototype was constructed and installed in the CHA Corporation laboratory. This prototype was modified numerous times as various operational problems were encountered. Task 3 work revolved around investigating process and engineering parameters and using that data to improve the prototype developed. Variable optimizations were based on a four stage breakdown of the diesel gas cleanup process. The first stage entailed analysis of the soot destruction parameters. The second involved adsorbent choice. Third, variable influencing adsorption efficiency was evaluated. Finally, parameters governing the regeneration of the selected carbon based adsorbent were analyzed.

After numerous modifications, the prototype design was finalized. The final prototype was used in the Task 9 demonstration in Sacramento, California in March 1998 (Task 9).

2.1.1 Soot Destruction

The intended goal of soot destruction testing was to maximize the decomposition of NOx while oxidizing filtered soot in a microwave field. Additional concerns were minimizing pressure drop and energy consumption. Soot destruction is completed through an in-line soot filter in which microwaves periodically induce combustion of the accumulated soot. The CHA Process provides additional heat and molecular excitation by applying a high energy microwave field to the filtered soot. Soot efficiently absorbs microwave energy and is quickly heated. Several filter configurations were assembled and tested. The two primary filters were commercially available ceramic monoliths manufactured by Corning, Inc and a wound filter made by the 3M Company. The ceramic filter performed best primarily because of low pressure drop during loading. The ceramic filter was incorporated in the final prototype.

2.1.2 Adsorbent Choice

Adsorption is the result of intermolecular forces (primarily Van der Waals) and is similar in effect to vapor condensation. Adsorption occurs at the gas-solid interface. As a result, high adsorbent surface area is critical to adsorbent performance. Carbon based adsorbents are produced by partially combusting a carbonaceous material under conditions that allow uniform reaction. When the solid carbon oxidizes to form carbon dioxide gas, a void is left in the space where the carbon molecule had formerly resided.

Adsorbent performance significantly influences overall process efficiency. Traditional adsorbent production practices are slow and costly. Uniform reaction requires uniform heating of the carbonaceous material. This demands a slow temperature increase and heightened heat transfer through the application of steam or other medium. Consequently, time and energy demands make adsorbent generation and regeneration expensive using conventional methodology. Microwave fields, however, can be used to uniformly heat the carbonaceous material, allowing for rapid generation and regeneration of the adsorbent. As a result, the CHA process for diesel gas treatment employs carbon-based adsorbents and microwave energy to take advantage of this quick and cost effective technique.

In diesel generated exhaust gas, NOx is present mostly as NO. An important aspect of NOx adsorption and destruction is that the NO must be in NO_2 form to adsorb on carbon within any reasonable operating temperatures[13]. The combined Van der Waals and polar attraction forces for NO are very small (NO boiling point is -241 °F (-152 °C) vs. 70 °F (21 °C) for NO_2). Therefore, conversion of NO to NO_2 is critical to pollutant removal. At process temperatures, the conversion reaction maintains equal quantities of NO and NO_2 unless the NO_2 is continuously removed.

Adsorbent Particle Size Testing

Four different carbon products, crushed form coke briquettes, 10-mesh calcine char, granulated activated carbon (GAC) 8/16 mesh, and GAC pellets, have been tested. Each of these adsorbent materials has a different grain size. With each of the carbon products, the minimum residence time is in the range of 2 seconds with little variation between products. Because of a high pressure drop, adsorbent size less than 10-mesh is not suitable.

Adsorbent Testing Results

The NOx breakthrough testing results suggested two possible adsorbents for use in the NOx adsorption column. Granulated Activated Carbon (8/16) and the GAC pellets performed more effectively than the other sample types. To obtain the NOx adsorption data, a 2-inch (5 cm) ID with 24-inch (61 cm) bed height column was used. The diesel exhaust gas was passed through the column at 35 SCFH (60 sm^3/h). The following table describes the respective performances.

Table 2: Adsorbent Performance Comparison

Adsorbent	Size	NOx Inlet Average (ppm)	20% Breakthrough Time (hrs)	Adsorption Capacity (g NO/100g C)
FMC Form Coke	<1/4"	638	2.72	1.47
FMC Form Coke	10 mesh	622	2.16	0.51
FMC Calcined Char	10 mesh	661	4.03	1.39
FMC Calcined Char	10/20 mesh	719	0.85	0.77

The amount of NO adsorbed per 100 grams of carbon adsorbent (adsorption capacity) was calculated by integrating NO adsorption over the saturation period until 80% of inlet concentration was achieved. The capacity as indicated represents the number of grams of NO adsorbed per 100 grams of adsorbent. The time in Table 2 is the 20% breakthrough time indicates the time for the outlet gas NO concentration to reach 20% of the inlet NO concentration. The breakthrough curves are shown in Figures 2-5. The adsorption data clearly indicates that the GAC performs more effectively than the other carbon adsorbents.

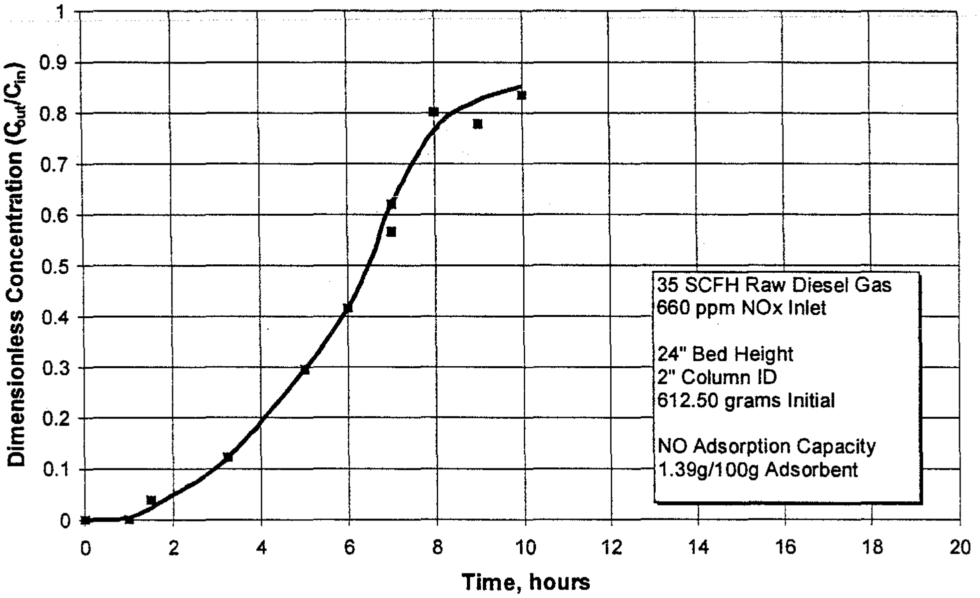


Figure 2: NO Breakthrough for 10 mesh Char

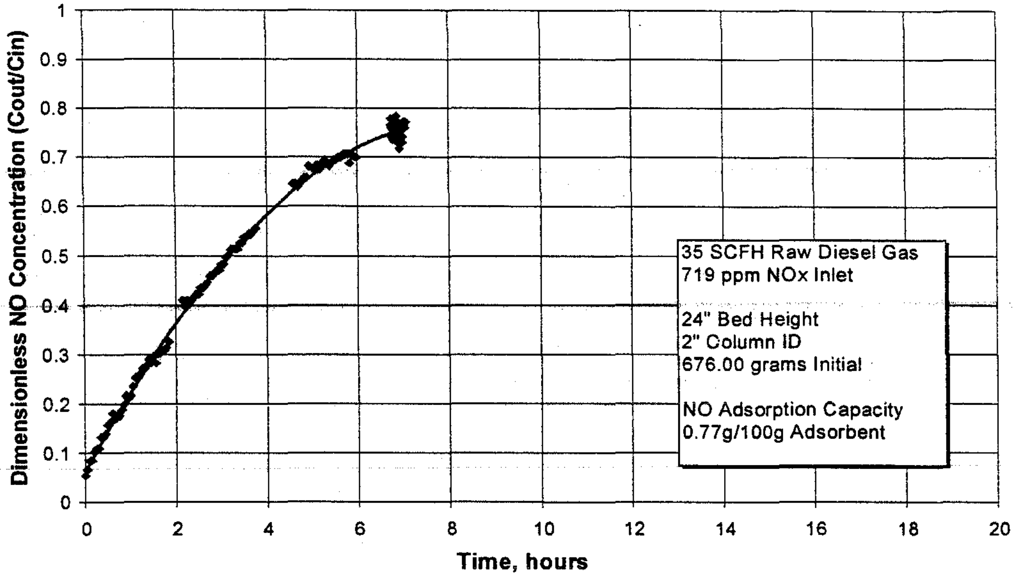


Figure 3: NO Breakthrough for 10/20 mesh Char

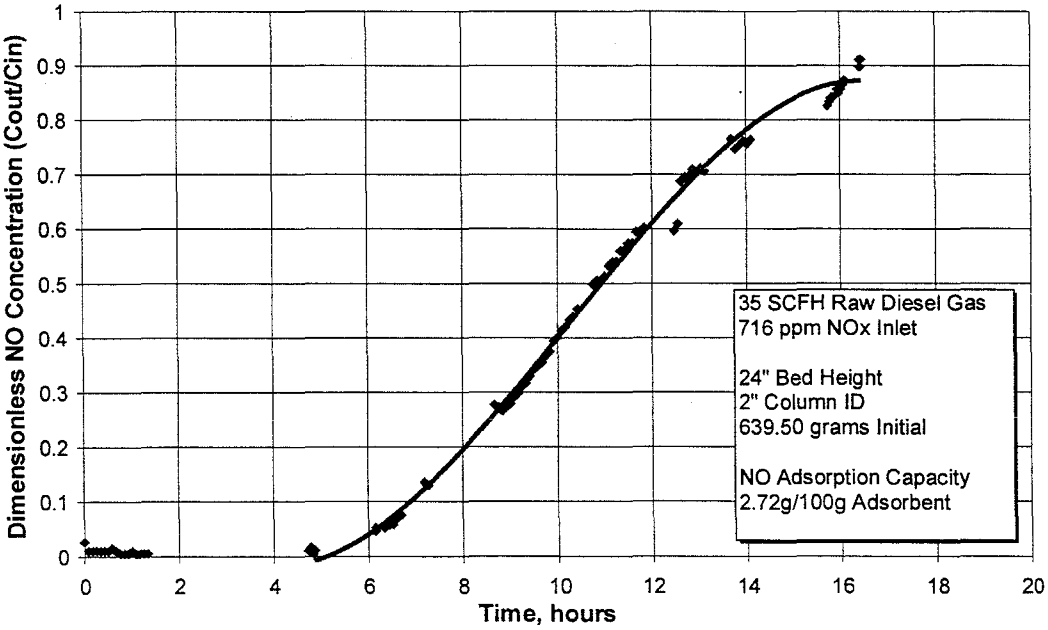


Figure 4: NO Breakthrough for GAC

In addition to the 2-inch adsorber, a large batch adsorber was used to evaluate the various adsorbents with 110 SCFM (187 sm^3/h) diesel exhaust gas. This adsorber is a tapered vertical cylinder 5 feet (1.5 m) tall and 30-inches (0.76 m) in diameters on the large end and 18-inches (0.46 m) in diameter in the small end. The following presents the NOx adsorption results obtained from the adsorber and moving-bed adsorber.

Crushed Calcined Char Briquettes. The FMC Corporation in Kemmerer, WY produces 1-1/2 inch (3.6 cm) briquettes of calcined char bonded with polymerized coal bitumen. NOx breakthrough was almost immediate and total NOx capacity was poor at less than one percent by weight. Significant by-passing and the immediate breakthrough at 200 ppm NOx was also noted with a residence time of 1.9 seconds. Pressure drop was very good at 3 inches water for the 38-inch (0.96 meter) bed used (large adsorber) with 25 ft/min (0.13 m/s) gas velocity. Near complete saturation of the char bed occurred within 4 hours where the outlet NOx concentration exceeded 700 ppm. It was determined that residence times as high as 3 seconds did not improve the char's NOx adsorbing performance. We also found that the 1/2-inch (1.3 cm) material was difficult to regenerate. Microwave power, 1500 Watts, applied to the crushed briquettes passing through the regeneration reactor at 30-35 lbs/hr (13.6-15.9 kg/hr) required 4 passes to regain approximately half of the NOx capacity measured in the original material. Attrition percentage was very low at 0.5% per pneumatic cycle.

10-mesh Calcined Char. The calcined char is the carbonaceous feed stock taken before briquetting. Therefore, it is free of the bituminous binder. The surface area of the calcined char is between 100-150 m^2/g, as opposed to the briquettes that have a surface area of approximately 250 m^2/g. The 10-mesh char was placed in the same large (18-36 inch diameter, 45-91 cm) absorber and the bed height was 36 inches (0.91 m). Initial NO breakthrough was observed within one hour and the bed was completely saturated within 6 hours. Microwave regeneration was achieved at 15 lbs/hr (6.8 kg/hr) and 1200 Watts of applied power. However, the capacity of the char to absorb NO is low at less than 2% by-weight absorption capacity. A residence time of up to 3 seconds did not significantly improve the char performance. The attrition rate of the calcined char was approximately 1%.

Granular Activated Carbon. Calgon F 6-10 granulated activated carbon (GAC) is a commercially available activated carbon made from sub-bituminous coal specifically for automotive applications. The adsorber bed height was 32 inches (81 cm). Breakthrough was observed in eight hours and good NO capacity was measured at 6% by weight. In addition, the GAC regenerated easily with 600 Watts applied microwave power at a feed rate of 15 lbs/ hr (6.8 kg/hr) through the reactor. Complete NOx saturation was observed after 36 hours of run time. This material was the most favorable for NOx removal of any material used. However, the mechanical strength of the GAC is significantly less than the char products. A large amount of fines was produced when used in the moving-bed reactor. The attrition rate was 5% per transfer and the fines were very difficult to remove from the pneumatic vents because of the fine powder consistency of the fines.

Granulated Activated Carbon Pellets. The final material tested was from the Filtration Media Group in Union Gap, Washington. We received 750 lbs (341 kg) of activated carbon pellets 4-mm in diameter and 8-10 mm in length. These pellets have a surface area of 1000 m^2/g. Attrition is less than 0.1%. After one hour of NOx absorption testing, there is no measurable NOx leaving the 38-inch (0.97 m) bed. The NOx adsorption capacity is very close to that of GAC. Due to the high adsorption capacity and low attrition rate, we chose the pellets for use in further prototype applications. In addition, NOx is easily desorbed, instead of reacting, with the carbon so that the activated carbon is not consumed.

In order to study the adsorption of a continuous moving bed, a 5-inch (12.8 cm) moving bed system was constructed and is shown in Figure 5.

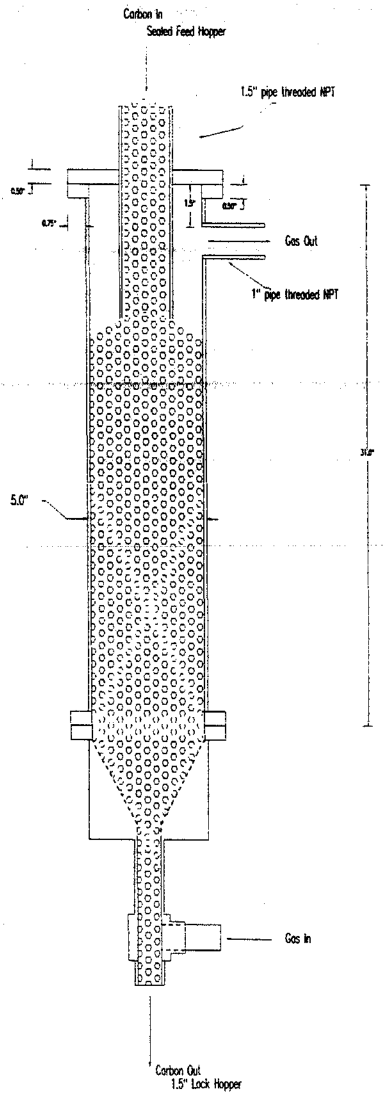


Figure 5: Moving Bed Design

The results of the moving bed adsorption efficiency testing with granulated activated carbon are depicted in Figure 6. The data is displayed with data point symbols differentiating bed height and best fit line shading corresponding to diesel exhaust flow rate. These results indicate the expected trend of improved efficiency with increased dimensionless mass flow rate. The dimensionless mass flow rate is calculated by dividing the rate of carbon adsorbent repletion in grams of carbon per hour by the NO throughput, also in grams per hour. This dimensionless group allows for scalable comparisons when considering larger column designs.

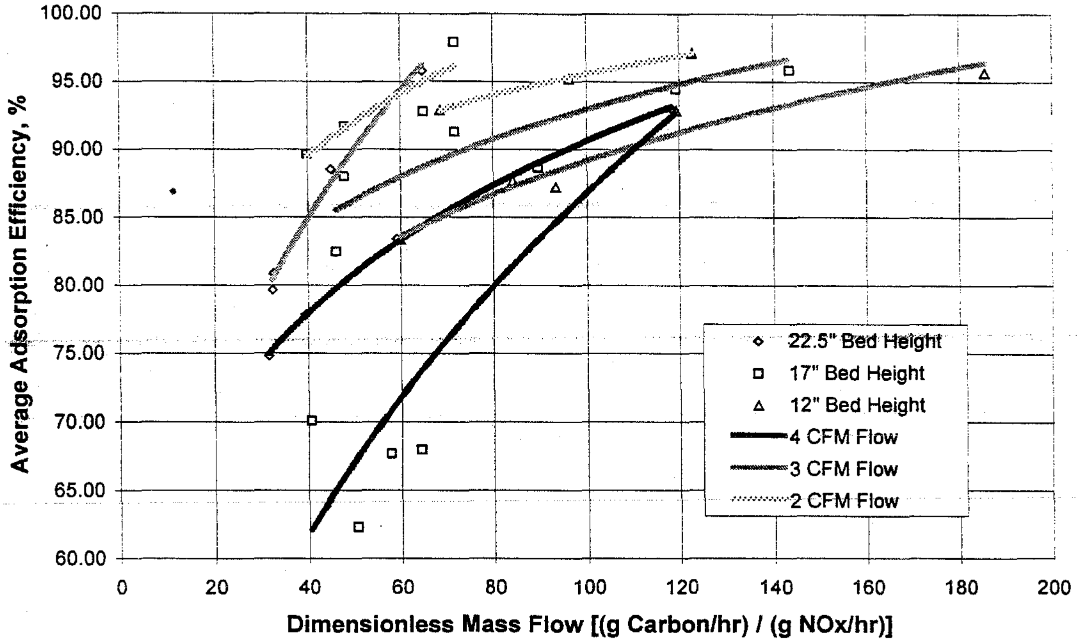


Figure 6: 5-inch Moving Bed Adsorption Efficiency for GAC

Results indicate that the efficiency of the moving bed column, assuming a reasonable adsorbent repletion rate for a given exhaust flow rate, is independent of bed height as long as a minimum bed height is achieved. The results obtained from the 5-inch (12.7 cm) moving bed reactor were used to design a moving-bed for the prototype system.

2.1.3 Regeneration

Carbon based adsorbent regeneration has conventionally been performed by heating the adsorbent using hot nitrogen purge gas or steam until all pollutant molecules desorbed. The inert gas must be used to avoid combustion of the carbon. Desorbed molecules are then destroyed or processed in a secondary reactor. In the CHA process, the regeneration procedure employs microwaves to remove and destroy the adsorbed pollutants in either one step or two steps.

When NO_2 adsorbed on char is exposed to a microwave field, sufficient energy is available for desorption in the same manner as conventional heating. However, the microwave field also provides sufficient energy to trigger parallel NO_2 - carbon reactions of the form:

$$NO_2 + C \rightarrow {}^1/_2\, N_2 + CO_2$$

$$NO + C \rightarrow {}^1/_2\, N_2 + CO$$

Although these reactions are exothermic, they do not produce enough energy to be self sustaining, and are consequently not favored in conventional regeneration. This will reduce adsorbent load and improve process efficiency. VOCs and CO released from the adsorbent will flow through a microwave based catalytic oxidizer and then be converted to CO_2 and H_2O. If the microwave regeneration is performed under a nitrogen purge, carbon will be consumed on a one-to-one molar basis with NO_2 destruction. The main problem with one step regeneration processes is the consumption of carbon adsorbent. NOx adsorbed in the activated carbon is easily desorbed during the microwave regeneration. The desorbed NOx flows into the second microwave catalyst reactor where NOx is decomposed into N_2 and O_2. The VOCs also react with NOx in this reactor.

As a result of meanderline difficulties with the original prototype, a continuous regeneration T-reactor was designed. The T-reactor uses timer actuated lock hoppers to add and remove carbon adsorbent. As the carbon passes through the quartz tube it travels through the microwave field coupled through the helix surrounding the quartz tube reactor. By changing the actuation period and energy input, adsorbent regeneration rate can be equated to adsorption column carbon repletion demands. This allows for the easy integration of the moving bed adsorption column with the regeneration apparatus to form one continuous cycle. The helical T-reactor regeneration device is presented in Figure 7.

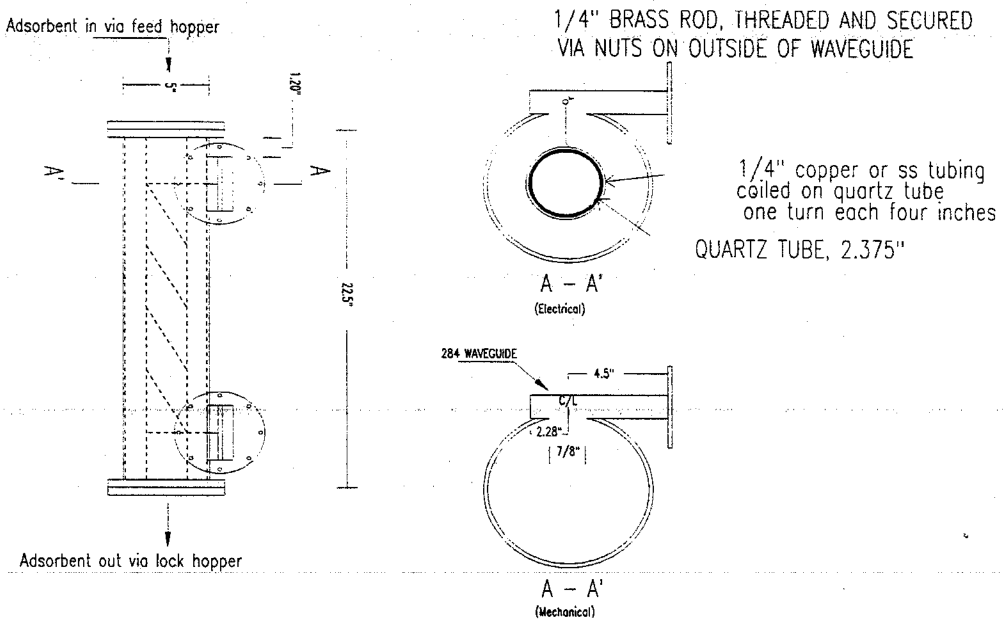


Figure 7: Helical Moving Bed T-Reactor

2.2 Test Soot Destruction Device (Task 5)

During Task 4, a soot destruction device was installed as part of the overall assembly. A series of tests were then carried out specifically aimed at the performance and regenerating ability of the soot destruction section.

After the filters were saturated with soot, they were exposed to between 750 and 900 Watts of microwave energy with an air flow rate of 4.5 SCFH (0.12 m^3/hr). Experimentation with both power levels was carried out by regenerating the filter off line with an air flow to support soot oxidation. The microwaves excite the small soot particles and create interparticular friction. This friction translates the microwave energy into heat energy providing the impetus for soot oxidation. As a result of the oxidation mechanism, sufficient soot accumulation is a requirement for successful regeneration.

Experimentation has shown that the ceramic monolith filter has a collection efficiency of approximately 90%. The ceramic monolith, wrapped directly with a helix, was easily regenerated. Regeneration of the ceramic monolith filter with microwave made the pressure drop across the filter return to the pressure drop prior to the saturation of the filter with soot. After completing the regeneration tests with the ceramic monolith filter wrapped directly with a helix, the microwave system was modified to reduce gas leakage.

Figure 8 shows a schematic of the new dual magnetron monolith design. This configuration has shown the best results so far. This device is six inches long and 5 5/8 inches (14 cm) in diameter and centered in the cavity with three inches of open space on either end. The two-inch magnetrons are placed directly over the ends of the cavity. The diesel exhaust enters the cavity and is passed through the filter end to end.

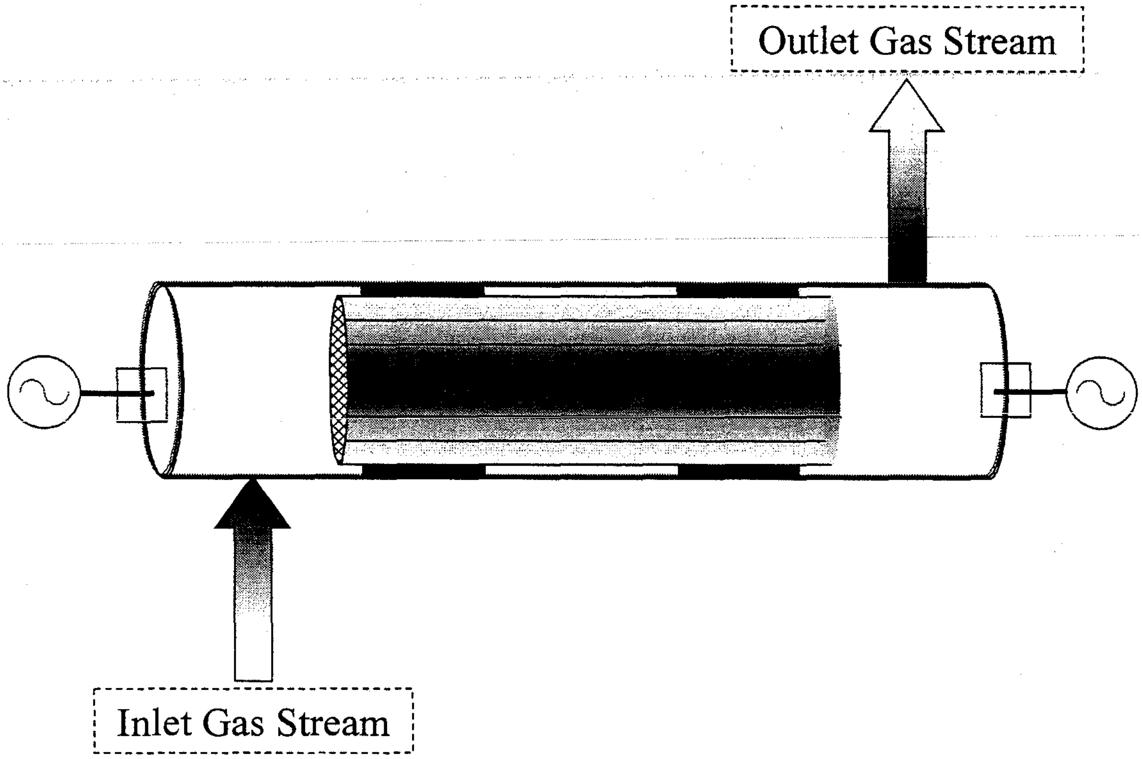


Figure 8: Ceramic Monolith Filter with Cavity Microwave Application

Over 50 regeneration tests have been conducted on this new ceramic monolith filter. The pressure drop across the filter returned to the original pressure drop after regeneration. This clearly demonstrates that soot deposits inside the filter were completely oxidized by microwave energy.

2.3 Fabrication of Regeneration Device, Build and Test Heat Exchanger and Investigation of NOx Adsorption

As a part of Tasks 6 and 7, construction of the heat exchanger and the overall system was completed and initial shakedown testing was completed.

Data representing several hours of operation are presented in Figures 9-12. Figure 9 shows the pressure drop across the monolith soot filters as a function of time. The periodic lowering of this pressure drop represents a regeneration cycle.

The adsorber was filled with 10-mesh FMC char for mechanical performance tests. Figure 10 illustrates NOx concentrations entering the bed and corresponding exit gas NOx concentration. The downward trend of the inlet NOx indicates that the bed is deteriorating. Figure 11 shows the NOx concentration as a function of time followed by Figure 12 which shows the NOx removal efficiency. These early data show only 50% removal with FMC char in the adsorber. The NOx removal efficiency will be improved when the char is replaced by GAC. No serious mechanical problems were encountered.

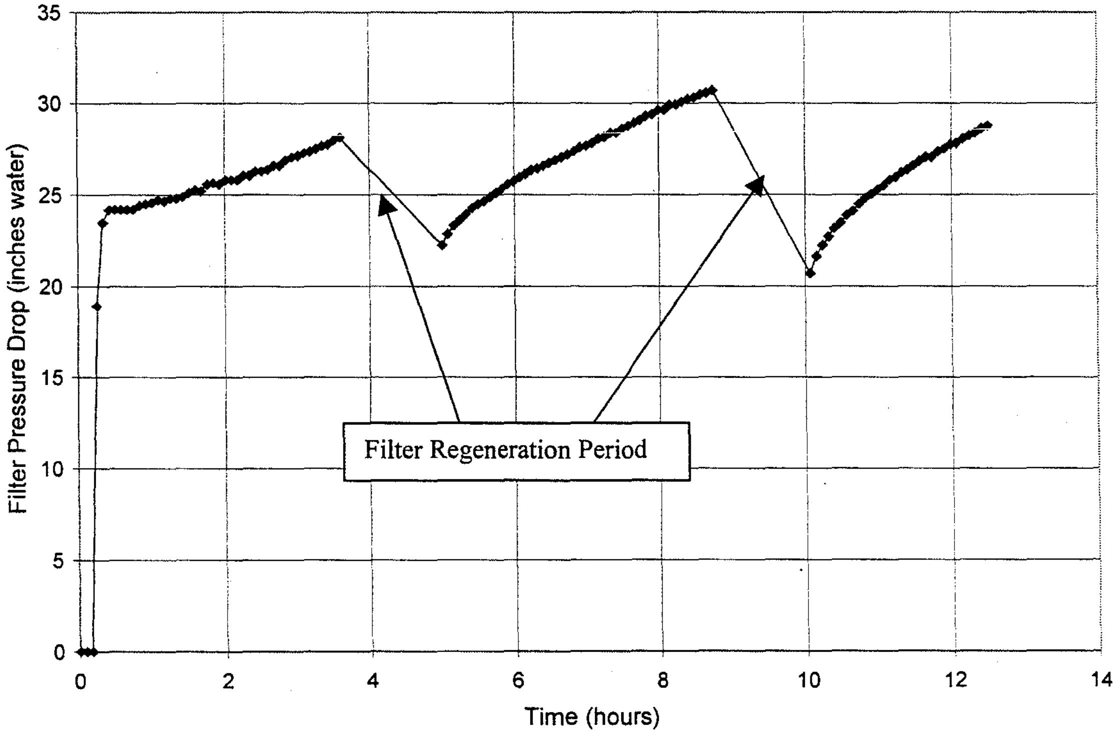


Figure 9: Pressure Drop Across the Soot Filter as a Function of Time

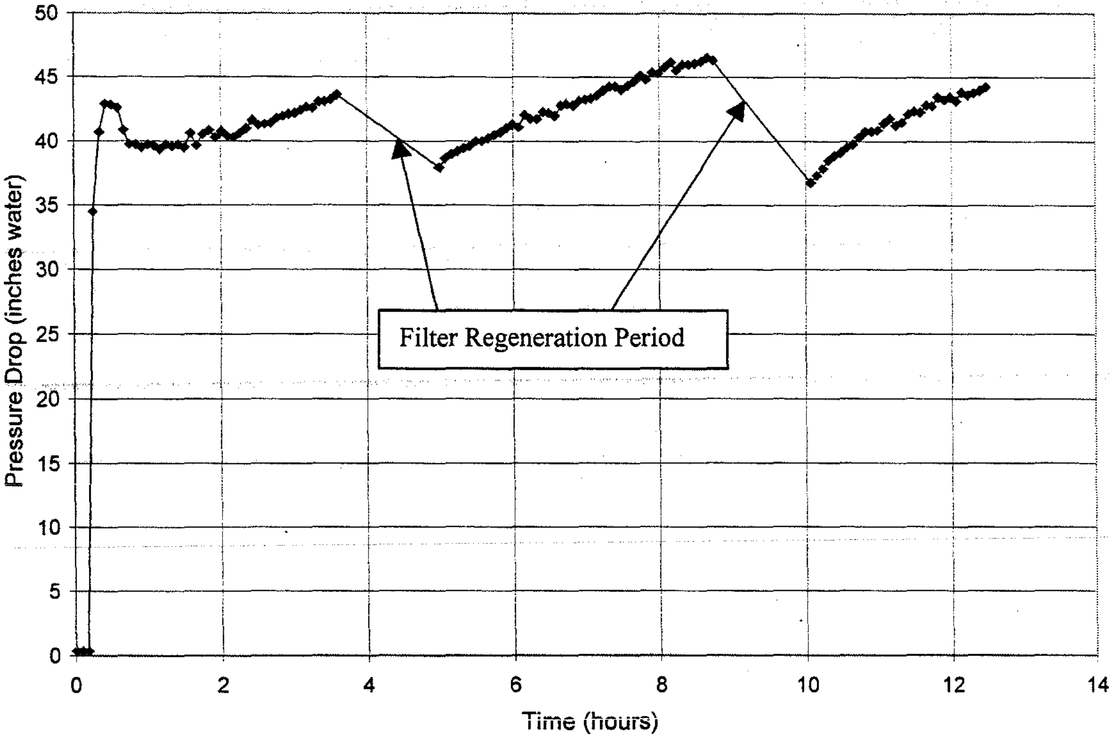


Figure 10: Pressure Drop Across the Engine as a Function of Time

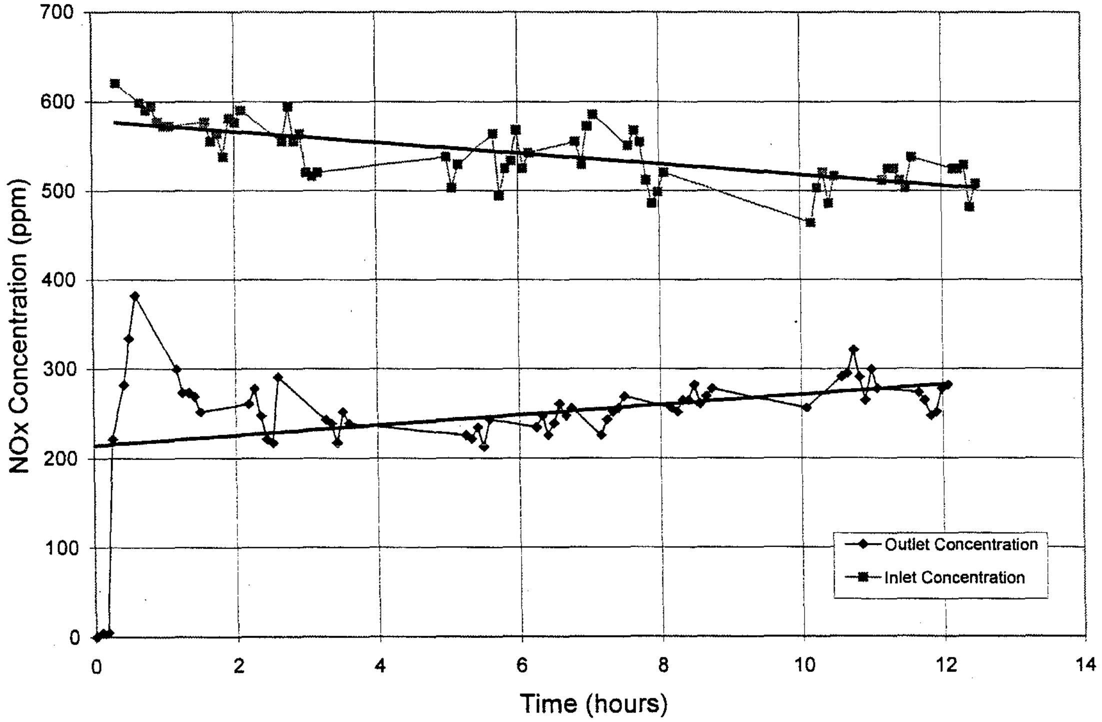


Figure 11: NOx Concentration as a Function of Time

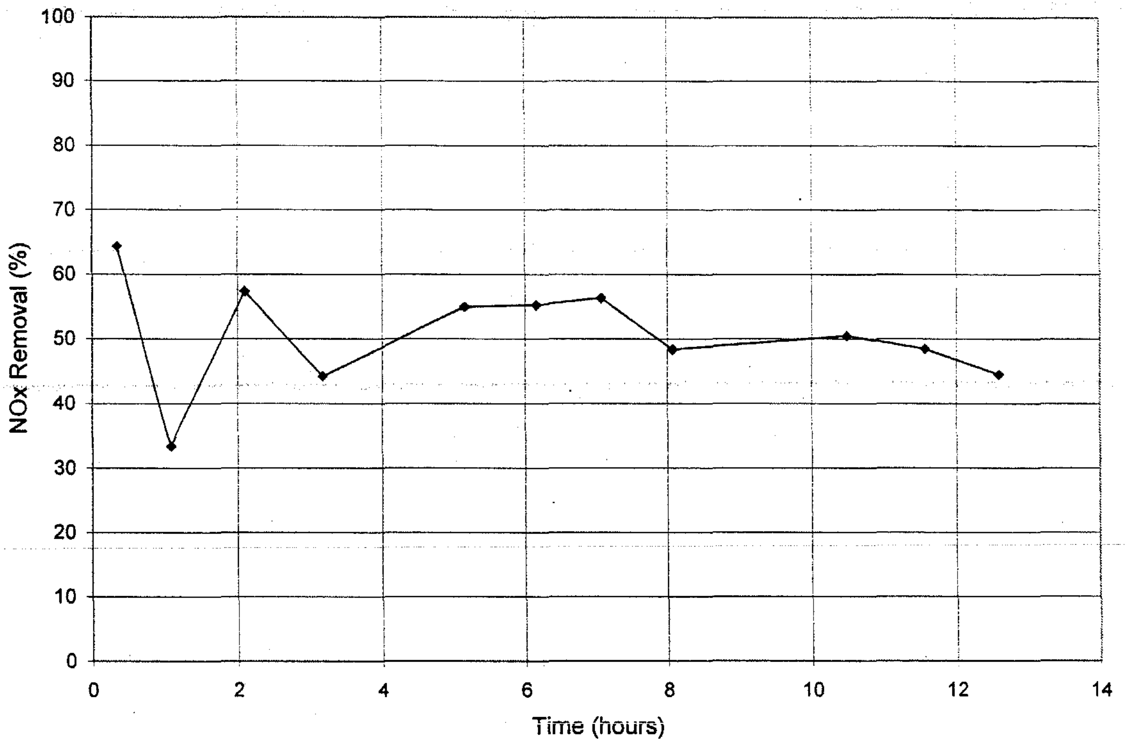


Figure 12: NOx Removal Efficiency as a Function of Time

2.4 Regeneration System

As a part of Task 8, we completed a number of long-term durability tests and refined various parts of the integrated system to accommodate operational problems observed.

The prototype system ran for periods of 24 to 48 hours to determine if operational problems were encountered. The changes made to the regeneration system are shown in Figure 13.

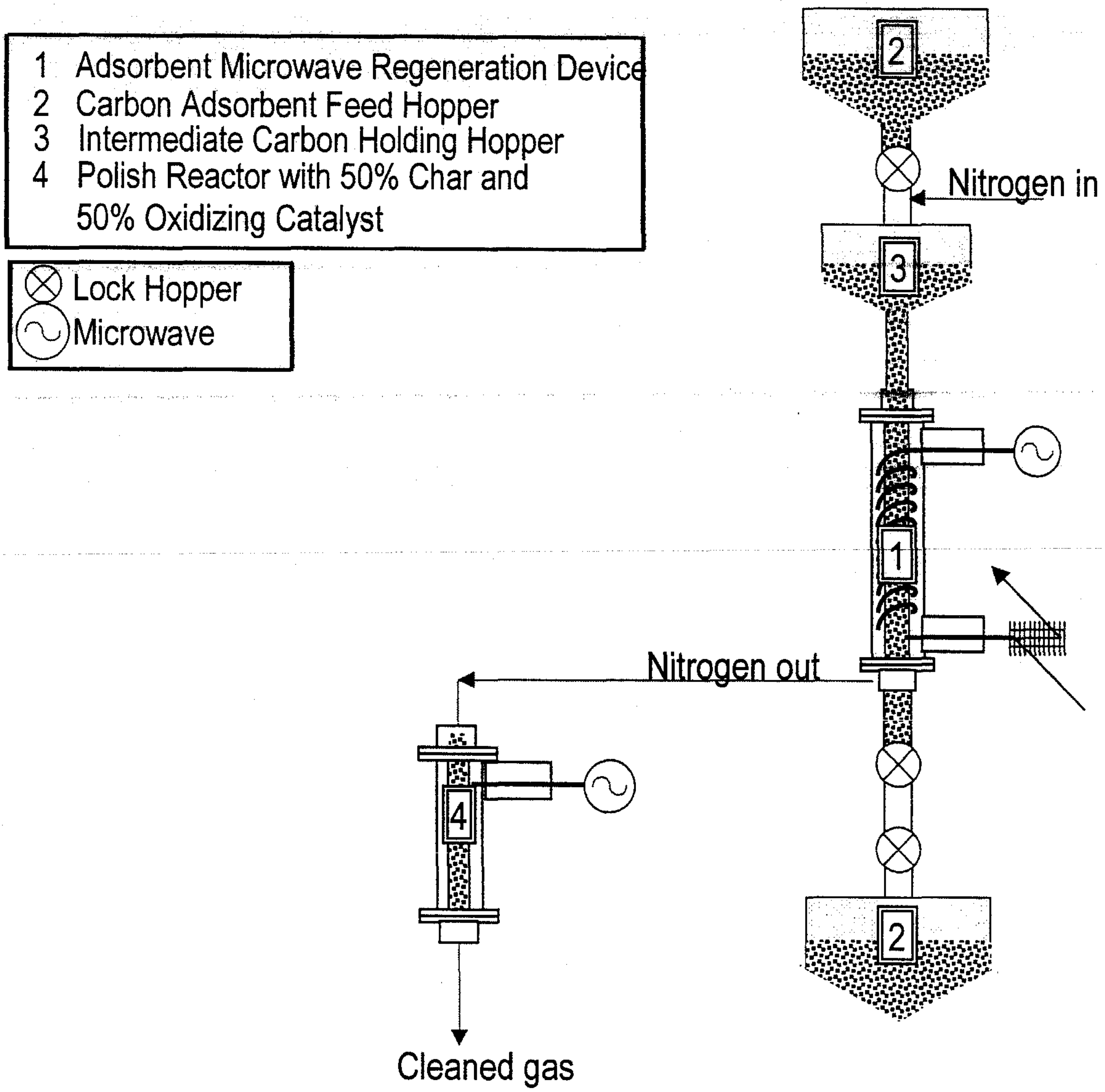


Figure 13: Newly Re-configured Regeneration Prototype

Figure 13 shows the changes made to the regeneration unit. Previously, the nitrogen purge gas was introduced at the top of the microwave regeneration device. The nitrogen gas, along with water vapor from the reactor, would escape into the carbon feed hopper (2) when the feed valve was opened, causing both the loss of the nitrogen and the contamination of the feed carbon with condensing water vapor. During long term run testing we discovered that the feed carbon would bridge occasionally, interrupting the flow of carbon into the reactor. To reduce the frequency of the feed valve operation, we added an intermediate feed hopper to store the feed carbon for a longer period of time between feed valve operations and moved the nitrogen addition point to the top of this new hopper (3). This change eliminated the feed carbon bridging problems. The nitrogen now continuously exits through the bottom of the microwave reactor and sweeps gas through the polish reactor. The polish reactor is packed with eight inches of calcined char and eight inches of oxidzing/reducing catalyst coated onto 1/8-inch (3.8 cm) alumina beads impregnated with 25% 800 mesh SiC.

2.5 Integrated CHA Diesel Exhaust Treatment Prototype System

Based upon the test data of each component, the prototype design was finalized. Figure 14 shows the finalized process diagram for the treatment of stationary diesel exhaust gas. The system represents a continuous process of adsorption and regeneration through which diesel exhaust gas can be continuously cleaned. The saturated carbon adsorbent is passed through a regeneration column where microwaves are used to regenerate the carbon. Diesel exhaust flows into the carbon adsorber at 150 °F (65.6 °C) and 110 CFM (187 m^3/hr). The activated carbon pellets, instead of GAC, were used as the carbon adsorbent because of its mechanical strength. The activated carbon pellets generated negligible fines during transport. As is apparent in the schematic, diesel exhaust gas first encounters an oxidation catalyst converter that oxidizes and destroys more than 90% percent of the VOCs and CO in the exhaust gas. The gas then flows into a microwave regenerated soot filter where more than 90 percent of the soot is removed. The filter is periodically regenerated. The gas is then routed through a heat exchanger for cooling. The gas is cooled from a temperature of 500 °F (260 °C) to a temperature between 150-200 °F (65.6-93.3 °C) to better facilitate the adsorption of NOx onto the carbon. The gas enters into the activated carbon adsorber where the NOx is adsorbed onto the carbon. The clean gas then exits through the top of the adsorber. The saturated adsorbent passes into a hopper and is then introduced into the 2.31-inch (6 cm) ID regeneration column. In the regeneration reactor, it is purged with a nitrogen stream to prevent adsorbent oxidation. In the regeneration reactor, the NOx laden carbon is exposed to 2450 MHz microwaves under a nitrogen purge to desorb the NOx and regenerate the carbon adsorbent. The concentrated NOx stream flows into a oxidation/reduction catalyst bed where the NOx is destroyed. The regenerated adsorbent is then passed into a hopper and cycles back into the adsorbent bed.

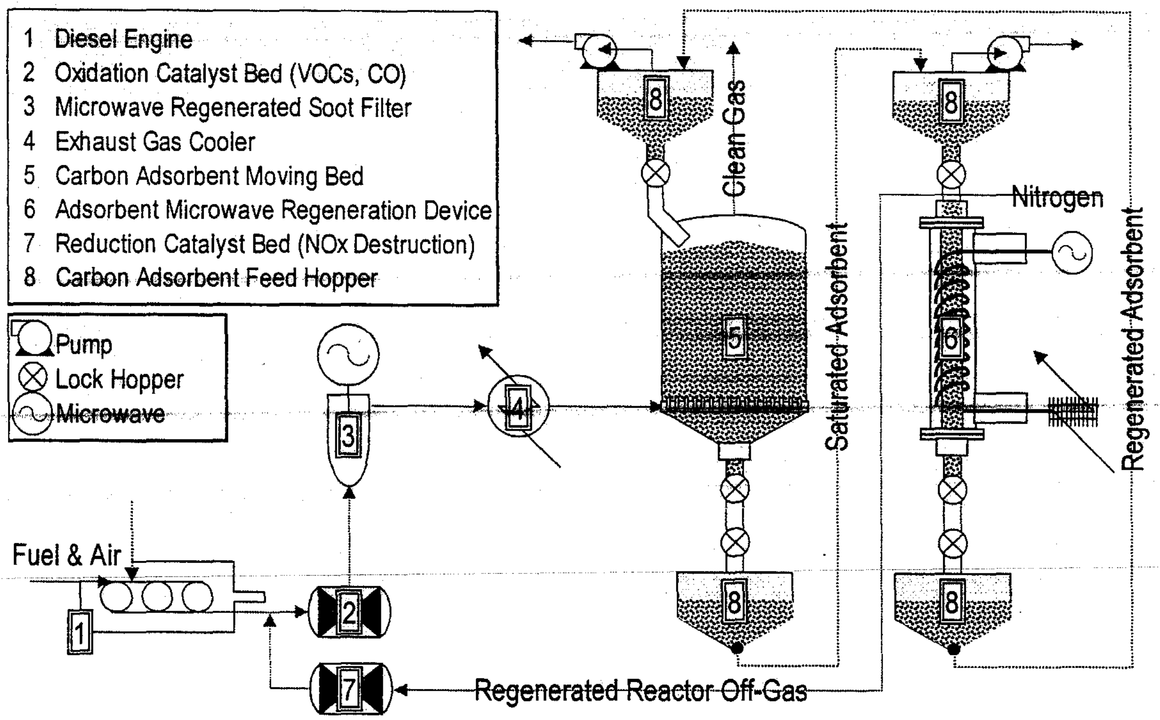


Figure 14: CHA Diesel Exhaust Treatment Process Flow Diagram for Prototype Demonstration

The updated integrated prototype design shown in Figure 14 is based upon suggestions by McClellan Air Force Base personnel to produce a continuously regenerated moving bed system that would allow for uninterrupted operation. The various prototype components are described below:

1. Diesel Engine- the diesel engine is a 58 hp, naturally aspirated diesel test engine. It operates at 1800 rpms and generates approximately 110 CFM (187 m^3/hr) exhaust gas. It is coupled to a 3 phase electric generator to which a load can be applied through a resistive heat bed. Exhaust gas is sent through the wall of the laboratory to where the integrated prototype system is assembled.

2. Oxidation Catalyst Converter- the oxidation catalyst bed is attached directly to the diesel engine manifold during prototype testing. It contains a ceramic monolith coated with an oxidation catalyst. Testing has shown that the catalyst effectively destroys over 90% of the CO present in the engine exhaust. This catalyst device is also effective at reducing effluent VOCs.

3. Microwave Regenerated Soot Filter- the microwave soot filter apparatus is the design used for testing the monolith soot destruction filters discussed in the previous section. The apparatus houses a ceramic monolith coated with an oxidation catalyst. Magnetrons at either end of the device introduce the microwaves into the monolith cavity. The 2.45 GHz magnetrons typically generate 800 Watts of microwave power. Cooling fans are attached to each magnetron to prevent overheating due to reflected microwave power.

4. Exhaust Gas Cooler- the exhaust gas cooler was designed to cool 120 CFM (201 m^3/hr) of gas from 500 °F to 100 °F (260-38 °C). The heat exchanger is equipped with a variable speed electric motor to control cooling capacity of the heat exchanger. The exchanger is outfitted with 2-inch (5 cm) NPT inlet and outlet gas fittings. The unit is made of aluminum and is approximately 2 feet by 2 feet (5 cm by 5 cm) and stands 3 feet (0.91 m) tall.

5. Carbon Adsorbent Moving Bed- the carbon adsorbent moving bed is 30 inches (0.76 m) square and 40 inches (1.01 m) tall. The carbon bed within the adsorption unit is 29 inches (0.74 m) tall. The gas is introduced through two 2-inch (5 cm) NPT inlet manifolds. The carbon is continuously fed through the adsorption unit via a feed hopper at the top and exits through a lock hopper at the base.

6. Adsorbent Microwave Regeneration Device- the adsorbent regeneration device has a 5 inch (12 cm) outer diameter and is 22 inches (56 cm) tall. The helix equipped cylinder houses a 2.31-inch (6 cm) ID quartz tube which is extended the full length of the interior of the reactor. Carbon is fed by a feed hopper at the top and exits by lock hopper actuation at the base. The carbon is transferred between the two systems by means of two pneumatic conveyors, which are periodically operated to transfer the carbon when the receiving hopper is full.

7. Microwave Reduction Catalyst Reactor- the microwave reduction catalyst reactor is a 2.31-inch (6cm) ID quartz tube packed with 8-inches (20 cm) of calcined char and 8-inches (20 cm) of oxidizing/reducing catalyst coated onto 1/8-inch (3.8 cm) alumina beads impregnated with 25% 800 mesh SiC. The char and catalyst work together to decompose any NOx leaving the main reactor.

3.0 Results and Discussion

A prototype unit for treating 110 SCFM (187 m^3/hr) of diesel exhaust gas produced from a 35 kW (58 hp) diesel engine was successfully designed, assembled, refined and demonstrated during this study. The prototype unit is the first one of it's kind to utilize the CHA NOx removal process to continuously remove NOx and VOCs from diesel exhaust gas. A soot removal feature was also developed and added to the prototype system to protect a heat exchanger from plugging. Prior to this study, the CHA NOx removal process had been tested on a lab scale on a batch basis. NOx and VOCs were adsorbed onto a carbon adsorbent and after the adsorption step the carbon was removed from the contactor and regenerated using microwave energy. Once regenerated, the carbon was returned to the contactor for re-saturation. During this study, the CHA NOx removal process was scaled up from a few cubic feet per minute lab scale to 110 CFM (187 m^3/hr) and the various process steps were integrated into an automated prototype, tested and improved to a point where the prototype unit could be demonstrated. The demonstration was successfully carried out in Sacramento, CA. during March, 1998.

This study provided an opportunity to incorporate the CHA NOx removal process into an integrated continuous prototype system. This prototype system is automated and operates continuously without operator input. The carbon is held in a moving bed adsorber where exhaust gas is passed through the carbon bed counter current to the flow of carbon. After passing through the moving bed, the NOx and VOC laden carbon adsorbent is automatically transferred to a moving bed microwave regeneration reactor where the adsorbed NOx and VOCs are converted to harmless carbon dioxide, nitrogen and water. After regeneration, the carbon is automatically returned to the moving bed adsorber.

The scale for the prototype system is 110 CFM (187 m^3/hr). This prototype unit was successfully demonstrated during March 1998 at McClellan Air Force Base. During the prototype demonstration, the engine used produced between 1000-1100 ppm of NOx. After passing through the CHA integrated prototype system, the outlet NOx concentration was 0 ppm. An account of the demonstration follows.

3.1 On-Site Demonstration

On 27 February 1998, two representatives from CHA Corporation, Charlie Carlisle and Bob Guffey, traveled to Sacramento with the CHA diesel exhaust treatment prototype in order to carry out an on-site demonstration at McClellan Air Force Base. By the end of Monday, 2 March, the demo unit was completely assembled and ready for testing. A 35 kW diesel generator was positioned near the prototype unit and piping connections were made. On Tuesday, 3 March, a load bank was connected to the generator set and a series of shakedown runs were completed. During the initial shakedown period, some concern arose because the test engine was in very poor condition and the exhaust gas contained an unusually high concentration of soot. The pressure drop across the soot filter unit elevated from 14 inches H_2O to 45 inches H_2O in 20 minutes of loaded run time because of the excess soot. This was unusual because typically, the engine start pressure was 10 inches H_2O with an elevation to 24 inches H_2O after eight hours of loaded run time. The McClellan 35 kW unit was powered by a six cylinder White engine that produced 73 hp and 148 CFM (252 m^3/hr) of exhaust gas. The McClellan engine produced about 40 CFM (68 m^3/hr) more gas than the 58 hp, 35 kW test engine used at CHA Corporation. In order to proceed with demonstration runs, one of the two filter elements in the filter unit was removed.

Also on Wednesday, a newer generator set was moved in and attached to the demo unit. The first engine used was worn out significantly and the soot volume was unusually high. The maximum NOx concentration in the exhaust gas of the first engine was only 375 ppm. The newer engine had fewer hours of run time and generated considerably less soot. The NOx concentration in the exhaust gas of the newer engine was between 1000 and 1100 ppm when loaded. The balance of runs was carried out using the newer generator set. Runs were continued throughout Wednesday, Thursday and Friday. The total run time on the unit was 9.2 hours. The outlet gas did not contain any measurable NOx indicating that essentially 100% of the NOx was removed in the adsorber. Wednesday, 4 March was the first day that the prototype was demonstrated for various visitors. The following is typical test conditions for the demonstration runs:

Typical Conditions for Test Runs

Exhaust gas temperature at filters	650 °F (343 °C)
Exhaust gas temperature after cooler	137 °F (58 °C)
Gas flow rate from engine	147 CFM (250 m^3/hr)
Engine Load	100%
Exhaust gas NOx concentration	1040 ppm
NOx concentration leaving unit	zero ppm
Engine manifold pressure	40 inches H_2O
Test room temperature	65 °F (18 °C)
Carbon feed rate	15 lbs/hr (6.8 kg/hr)
Regeneration microwave power	1600 Watts

During the demonstration, 100% of the NOx was removed from the test engine exhaust gas. The prototype unit was operated about 9.2 hours. During the demonstration, the prototype unit was powered by electrical outlets in the test facility. In the field, the NOx abatement system will be powered by the generator unit being treated The activated carbon pellets used during the demonstration was new and had experienced very little exposure to NOx before the demonstration. After returning from the demonstration, the prototype system was set up in our laboratories and durability testing was continued using the same carbon. Figure 15 shows a NOx breakthrough curve that was taken during this durability testing period. The carbon bed was left stationary and regeneration was not operated during the first 11 hours of operation. This fixed bed NOx saturation period allowed us to see NOx breakthrough as a function of time for the activated carbon pellets. After 11 hours of exposure to the 110 CFM (187 m^3/hr) the capacity of the carbon adsorbent had reduced to a point where only about 47% of the NOx was being removed form the diesel exhaust gas. At this point, the carbon handling system was started and the microwave regeneration system was turned on. This converted the adsorption bed to a moving bed system and the carbon started microwave regeneration at 19 lbs/hr (8.6 kg/hr). The NOx removal efficiency of the system improved within a short period even though the exhaust gas temperature was allowed to increase to 190 to 200 degrees F. NOx removal elevated from 47% to 74% within three hours. This NOx removal improvement shows that the microwave regeneration system is more than adequate to handle the design flow rate of 110 CFM (187 m^3/hr). Following this series of tests, represented in Figure 15, the exhaust gas flow was stopped and the regeneration system continued to run until the carbon bed was completely replaced with regenerated carbon. The regeneration of saturated activated carbon in the adsorber required 37 hours.

Exhaust gas flow was then resumed at 110 CFM (187 m^3/hr), and the data shown in Figure 16 was recorded. These data show that 80% of the NOx was continuously removed at 140 degrees F. This test also showed that after total regeneration, the carbon NOx adsorption capacity was reduced at least 20%. In earlier tests, such as the demonstration, NOx removal efficiency was 100% under the same conditions. This 20% decrease in capacity is typical of activated carbon applications. The initial, new carbon, capacity is generally referred to as virgin carbon capacity and after regeneration the carbon adsorption capacity is referred to as working capacity.

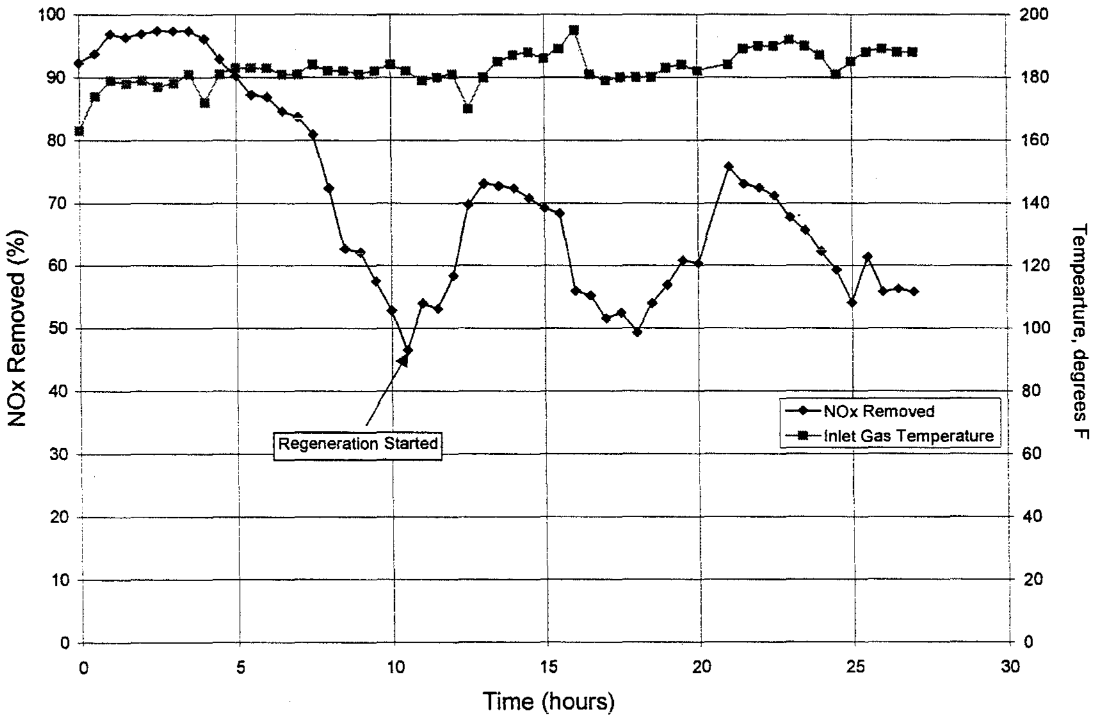


Figure 15: Bed Saturation Followed by Regeneration

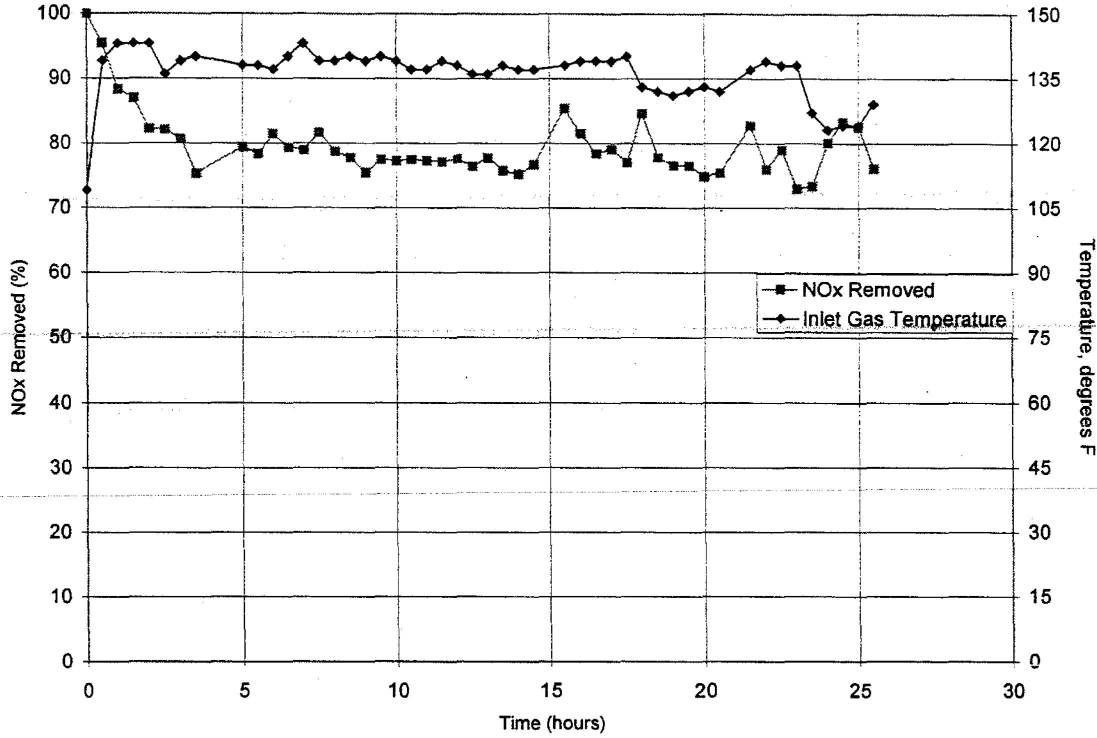


Figure 16: Steady State Operation

The variation of NOx removed after 15 hours, shown on Figure 16, is caused by poor control of the adsorption bed temperature. To confirm this observed loss in NOx adsorption capacity for the activated carbon a series of small adsorber NOx adsorption breakthrough tests were performed. In these test, a 2" (5.08 cm) by 24" (61 cm) adsorber was filled with fresh activated carbon pellets identical to the ones used in the demonstration tests and a mixture of NOx at 1000 ppm, and air diluted with nitrogen to achieve a 14% oxygen concentration, flowing at 0.55 CFM (0.93 m^3/hr) was passed through the bed at room temperature. These flow represent a 25 feet/min. (0.13 m/s cm/min) superficial gas flow rate in the bed. NOx concentration was continuously monitored at the carbon adsorber bed inlet and outlet ports. When the outlet NOx concentration was one half of the inlet NOx concentration, the gas flow was stopped and the carbon removed, weighed, regenerated with microwave energy, then returned to the bed for a new cycle of NOx saturation/regeneration. This procedure was repeated for five cycles. The data showing the carbon NOx adsorption capacity for each cycle is shown in Figure 17. Interestingly, these tests confirm the loss of NOx adsorption capacity after the first cycle. The fresh carbon NOx adsorption capacity was 7.1 g NO/ 100 g C, and the second through fifth cycle NOx adsorption capacity almost constant at 4.0 g NO/ 100 g C. This is consistent with other activated carbons used in industry and regenerated with conventional thermal methods. Fresh carbon has about twice the capacity as regenerated carbon. Regenerated carbon capacity is commonly referred to as working capacity and fresh carbon capacity is referred to as virgin carbon capacity. Equally, it is important to note that the carbon was not consumed during this cyclic work. The adsorbed NOx quickly de-sorbed from the carbon during regeneration without reacting with the carbon. The weight of the carbon bed after regeneration was the same as the starting weight before saturation. This is a favorable characteristic since no carbon loss means an extended overall life for the carbon bed.

In order to achieve a 90% NOx removal efficiency, it will be necessary to increase the activated carbon bed height from 38 inches (97 cm) to 48 inches (156 cm). The conventional GAC did not show the decline in adsorption capacity when it was regenerated with microwave. However, the activated carbon pellets are preferred for the moving bed adsorber because of its mechanical strength.

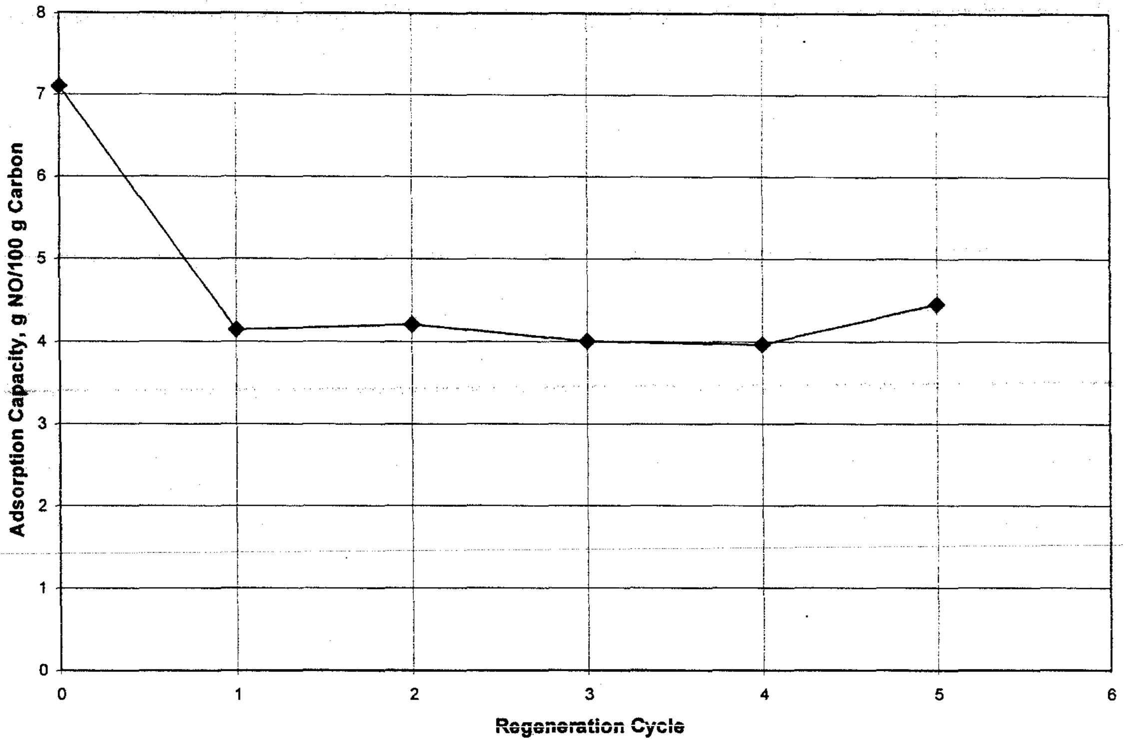


Figure 17: NOx Adsorption Capacity of 4 mm Activated Carbon Pellets

3.2 Economic Evaluation

We have estimated the capital and operating costs for a scaled up system which will accommodate the exhaust gases, 1,000 CFM (1700 m^3/hr), from a 250 kW (314 hp), diesel engine using the test data obtained from the 110 CFM (187 m^3/hr) prototype unit.

These data are based on steady state operating conditions consisting of an ambient temperature of 85°F (30 °C), a pressure of 28.38 inches Hg (0.95 atm), and No. 2 diesel fuel. In addition, the NOx shown is not present in the exhaust. It is based on the assumption that the NO is converted into NOx in the atmosphere.

An economic evaluation was prepared based upon components used in the prototype unit. In addition, operating costs were estimated for treating 1000 CFM (1700 m^3/hr) of diesel exhaust gas containing NOx, CO, VOCs, Hydrocarbons, and diesel particulate matter (DPM).

Exhaust gas produced from a diesel engine driving a 250 kW generator with a 100% load has the following composition (EPA Report # EPA-453/R-93-032, July 1993):

Exhaust Gas Composition from a Diesel Engine Driving a 250 kW Generator

	kg/hr	Volume %
Carbon Dioxide	158.57	8.14
Nitrogen	938.28	75.69
Oxygen	112.43	7.94
Water	62.74	7.87
Carbon Monoxide	0.91	0.08
NO	1.70	0.13
NOx	2.60	0.12
Sulfur Dioxide	0.20	0.02
Hydrocarbons	0.30	
Diesel Particulate Matter	0.012	

3.2.1 Capital Cost Estimate

The purchase cost for the adsorber was estimated by the method described in the textbook Air Pollution Control by David Cooper and F.C. Alley, pg. 441. The other equipment purchase costs were obtained from equipment venders and in-house cost data. The purchase cost for each equipment item is presented in Table 3.

Table 3: Estimated Equipment Costs

Equipment Name	Cost	Description
Adsorber	$13,300	5 feet diameter, 6 feet high
4-inch Lock Hopper Valves	3 x $1,000 = $3,000	Actuating valves
One Star Valve	$5,000	Continuous feed Star valve
Air cooler	$5,000	Heat exchanger used to cool the inlet diesel exhaust gas
4 hoppers	4 x $1000 = $4,000	Carbon holding device
Catalytic converter	$300	
Soot filter	$7,500	Monolith Soot Filter
Vacuum pump	$2,000	Used to transport carbon
Controller	$5,000	
Microwave generator and reactor system	$50,000 for 915 MHz or $24,000 for 2450 MHz microwave reactor system	Used for regeneration

The total estimated purchased equipment cost is $95,100 when a 915 MHz microwave generator is used for regeneration of saturated carbon. On the other hand, the total purchased equipment cost is reduced to $69,100 when 2450 MHz magnetrons and a conveyor belt microwave reactor for the regeneration are used. The vertical quartz reactor has been operated with 2450 MHz microwaves for a long time without any major problems. However, the penetration depth is only 2-inches (5 cm), which severely limits the use of a vertical quartz reactor. A 915 MHz microwave reactor will have a penetration depth almost three times greater. Consequently, a 10-inch quartz column can be used as a regeneration reactor if a 915 MHz microwave generator is used. This is the main reason for considering a 915 MHz microwave generator for the cost estimate. However, the capital cost is reduced by 27% if 2450 MHz magnetrons are used. A belt conveyor reactor has been constructed and tested using 2450 MHz microwave generator. Therefore, we have chosen a belt conveyor microwave reactor equipped with 2450 MHz magnetrons for regeneration of saturated carbon for capital cost estimate.

Using 5% freight and a 65% installation cost factor, the total installed equipment cost is \$117,470. The purchased activated carbon cost was estimated at \$4,310. Assuming 5% freight cost, the activated carbon adsorbent cost will be \$5,000. The total capital investment was estimated at \$122,470. This is equivalent to \$490/ kW.

3.2.2 Operating Cost Estimate

The plant is assumed to operate at the treatment rate of 1000 CFM (1700 m^3/hr) of diesel exhaust gas containing NOx, CO, VOCs, hydrocarbons, and dry particulate matter (DPM) for 365 days per year. It is also assumed that the plant is operated automatically and controlled by a computer. Hence, no personnel attendance will be required. Assuming that the labor equivalent of 1/4 person will be required to operate the plant, the loaded direct labor cost is \$13,000 per year.

Supplies and material costs, including the price of the activated carbon are estimated at \$20,000/year. The maintenance cost is assumed to be 3% (Air Pollution Control, a Design Approach, C. David Cooper, F. C. Alley, Waveland Press, Inc., 1994, Chapters 12 and 13.) of the capital investment, or \$3,675/year. The annual cost for taxes and insurance are taken as 3% of the capital investment, or \$3,675/year. The annual plant power demand is estimated at 65,700 kW*hr and the cost of power is estimated to be \$0.08 per kW*hr resulting in an annual cost of \$5,300. The total annual operating cost is \$45,650. This is equivalent to \$1,800/ ton of NOx or \$1,176/ ton of pollutants removed.

This compares to Selective Catalytic Reduction (SCR) cost which is about $1,800 per ton NOx removed for similar size installations. However, this comparison is not appropriate because SCR is not suitable for intermittent use such as the application discussed here.

However, the diesel engine is not used for 24 hours a day. The majority of diesel engines are used only approximately 175-2080 hours per year. Therefore, the annual operating cost will be $912 to $10,839/ year.

4.0 Summary and Conclusions

The purpose of this project was to design, fabricate, develop and demonstrate an integrated prototype system that would be capable of treating diesel engine exhaust for a 58 hp diesel engine. The work was broken down into nine tasks reported bi-monthly during the 18 month project. This integrated system is based upon a process founded by Dr. C.Y Cha in the late 1980's. Follow-up exploratory work was performed under the sponsorship of the United States Department of Energy (DOE) under Phase I and Phase II SBIR grants (DOE contract number DE-FG03-90ER80898) and was completed in August of 1993. The process is disclosed in four US patents (numbers 5,246,554; 5,256,265; 5,269,892 (process); 5,269,892 (equipment)). The integrated system is a continuous adsorption/ regeneration system capable of 100% NOx removal with adequate bed height, as well as destroying the VOCs and Soot present in diesel exhaust.

The process can be generally utilized on gas streams that contain NOx, SOx, or both NOx and SOx. The pollutants are first removed from the flue gases by passing the gases through a bed of relatively inexpensive carbon based adsorbent. The NOx, SOx and VOC gases are adsorbed and stored on the carbon during this step. The pollutants are then destroyed during regeneration of the carbon by using microwave energy. The microwave energy decomposes NOx to nitrogen and carbon dioxide, VOCs to carbon dioxide and water, and decomposes SOx to elemental sulfur and carbon dioxide.

The adaptation of the CHA process to diesel engine exhaust gas is a unique undertaking. The prototype system developed and demonstrated during this ICAT project is the first of its kind. Interestingly, applying the CHA process to diesel exhaust gas required the development of a diesel soot filter. This filter was required to protect the heat exchanger needed to cool the hot diesel gas, from plugging with diesel soot. The soot filter that was developed during this project, is a permanent filter with microwave regeneration capabilities.

The system represents a continuous process of adsorption and regeneration through which diesel exhaust gas can be continuously cleaned (Figure 14). The saturated carbon adsorbent is passed through a regeneration column where microwaves are used to regenerate the carbon. Diesel exhaust flows into the carbon adsorber at 150 °F (65.5 °C) and 110 CFM (187 m^3/hr). Diesel exhaust gas first encounters an oxidation catalyst bed that oxidizes and destroys almost 100 percent of the VOCs and converts CO to CO_2 in the exhaust gas. The gas then flows into a microwave regenerated soot filter where more than 90 percent of the soot is removed. The filter is periodically regenerated offline. The gas is then routed through a heat exchanger for cooling. The gas is cooled from a temperature of 500 °F (260 °C) to a temperature between 150-200 °F (65.5-93 °C) to better facilitate the adsorption of NOx onto the carbon. The gas enters into the activated carbon adsorber through two 2 inch (5 cm) pipe manifold distributors where the NOx is adsorbed onto the carbon. The clean gas then exits through the top of the adsorber. The saturated adsorbent periodically flows from the adsorber into a hopper and is then transferred to a 2.31 inch (6 cm) ID regeneration reactor. The regeneration reactor is continuously purged with a nitrogen stream to prevent adsorbent oxidation. Once in the regeneration reactor, the NOx laden carbon is exposed to 2450 MHz microwave energy under a nitrogen purge to desorb the NOx and regenerate the carbon adsorbent. The concentrated NOx stream flows from the regeneration reactor to an oxidation/reduction catalyst bed where the NOx is destroyed. The regenerated adsorbent is then transferred to a storage hopper. Periodically, the stored regenerated carbon is pneumatically transferred back to the adsorbent bed completing the cycle of carbon movement.

The CHA Diesel Exhaust Treatment System is based on the use of activated carbon and microwave energy. The prototype treatment system was designed, fabricated, operated and refined for the cleanup of diesel exhaust gas generated from a 58 hp diesel engine. The prototype unit was successfully demonstrated at the McClellan Air Force Base in Sacramento, CA. in March 1998. The prototype is a stand alone unit applicable to treating the diesel exhaust gases produced by stationary diesel engines such as standby generators. The prototype can easily be scaled up for use with diesel engines larger than 58 hp.

The ceramic monolith filter has a collection efficiency of approximately 90%. Two filters (5 5/8 inches in diameter and 6 inches long) were installed to the prototype unit. The pressure drop was acceptable for approximately 8 hours operation. After 8-hous of operation, the filters were successfully regenerated with microwave energy for over 50 cycles. For larger units, a 12-inch diameter monolith filter can be used. A magnetron, identical to those used in home microwave ovens, is installed in the monolith filter housing and is powered during regeneration to burn away the accumulated soot. After each regeneration with microwave energy, the pressure drop across the filter returned to approximately 10 inches of water which was the same pressure drop measured for a new monolith filter. Each filter element accommodates a flow rate of 60 CFM (102 m^3/hr).

The microwave regeneration of ceramic monolith filters can be completed within one hour. A combination of a bigger sized (a 12-inch diameter monolith is currently available) monolith and a larger number of filters can be easily adopted to treat a larger exhaust gas flow rate. The microwave-based filter has a strong possibility to be applied to underground mine diesel engines.

Both char and a variety of activated carbon products were tested for their ability to remove NOx from diesel exhaust gas and perform in the prototype system. During microwave regeneration, NOx reacts with char to produce CO and N_2, but easily de-sorbs from activated carbon. As a result, a small portion of the char is consumed during regeneration but the activated carbon is not consumed. Furthermore, the NOx removal efficiency is greater with the activated carbon than with char. Unfortunately, granular activated carbon is very soft and the fine particles generated during the transfer of the carbon from the contactor to the regeneration section or from the regeneration system to the contactor were difficult to control. Recently, we found activated carbon pellets, GAC, manufactured by the Filtration and Media Group, Inc of Union Gap, Washington. These carbon pellets are quite hard and produce a negligible amount of dust during handling. This carbon pellet was used for demonstration at the McClellan Air Force Base, and will be used in the first commercial applications.

A vertical adsorber is used for the prototype unit. This type of adsorber is applicable for gas flow rates up to 1000 CFM (1700 m^3/hr). For gas flow rates greater than 1000 CFM, a radial type adsorber should be used to reduce the size of the adsorber. Sorbent Control Technologies, Inc in Elgin, Illinois, designed and manufactures a radial adsorber that can be easily adapted for the NOx removal system.

The activated carbon pellets are capable of removing virtually 100% of the NOx in the diesel exhaust gas without problems associated with frequent shutdown and start up operations. In the prototype system, with a gas flow rate of 110 CFM, the bed height should be increased from the current 38" (97 cm) to 48" (122 cm) to achieve better than 90% NOx removal efficiency.

The saturated activated carbon was continuously regenerated in the microwave reactor at a rate of 17-20 lbs/hr (8-9 kg/hr). The NOx desorbed from the activated carbon pellets was decomposed to N_2 and O_2 by an oxidation/ reduction three way catalyst in a second microwave reactor. The catalyst contains silicon carbide that absorbs microwave energy and is coated with a platinum rhodium based catalyst. A 2.31-inch (6 cm) diameter quartz tube was utilized in the microwave reactor for the 110 CFM (187 m^3/hr) prototype unit. For a larger flow rate, a belt conveyor microwave reactor has been built and tested for the microwave regeneration of saturated carbon. A vertical moving bed reactor can be used for larger units if a 915 MHz microwave generator is used in place of 2450 MHz magnetrons. The use of magnetrons makes the microwave reactor much cheaper than using the microwave generators. Because of the cost and flexibility of using 2450 MHz magnetrons, a belt conveyor microwave reactor is recommended for larger units.

The maximum system pressure drop did not exceed 45-inches (114 cm) H_2O. The major pressure drop occurred at the soot filters (20-30 inches, 51-76 cm, H_2O). The exhaust gas cooler manufactured by Xchanger, Inc. in Hopkins, Minnesota worked well to cool 110 CFM (187 m^3/hr) of exhaust gas from 500 °F to 100 °F (260 °C to 38 °C). The heat exchanger is equipped with a variable speed electric motor driven fan to control the cooling capacity of the heat exchanger. The scale-up of the heat exchanger will not be difficult.

For the treatment of exhaust gas generated from a 250 kW standby electric generator, the capital cost was estimated at $122,470 or $490/ kW. The annual operating cost was estimated at $912 to $10,839 for 175 to 2080 operating hours per year. Assuming a 10% cost and a $14,000/ ton NOx removal credit, the minimum operating hours required to have a breakeven is estimated at 440 hours. If the system is operated for 8 hours every day, a $60,000 income can be generated from the NOx removal credit.

In conclusion, the prototype unit was successfully used to demonstrate the various steps of the CHA Diesel Exhaust Treatment System. The system can easily be scaled up to remove NOx and/or VOCs from the various gaseous effluents with reasonable economic return by taking a pollutant removal credit.

5.0 Recommendations

The prototype developed by CHA Corporation to treat diesel engine exhaust has proved very successful. Further research on scale-up procedures, long term durability and application to cleaning non-diesel gas streams is in progress. For successful commercialization of the process, it is recommended that a 1000 CFM commercial demonstration unit be constructed and field demonstrated.

The application of the prototype system is flexible and other gaseous such as VOCs for example, can be abated with slight equipment modification. It is recommended that a field demonstration of VOCs removal using the prototype developed during this study be carried out.

List of Inventions Reported and Publications Produced

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