INDIRECT MICROWAVE TREATMENT OF DIESEL EXHAUST GASES FOR NO_x CONTROL

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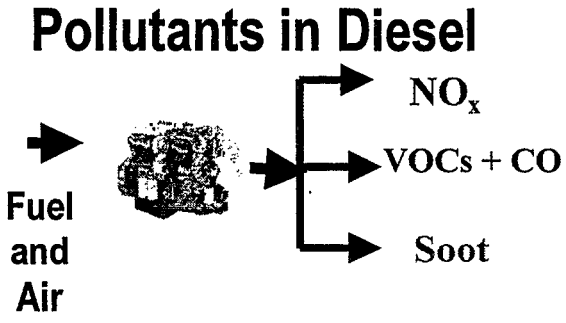
INTRODUCTION

As shown in Figure 1, diesel exhaust gases contain a number of combustion products that have attracted considerable attention during the past couple of decades. Soot and VOCs, generated by all diesel engines, have been linked to cancer and lung disease in humans[1]. Equally NO_x, emissions contribute to smog formation and acid-rain[2].

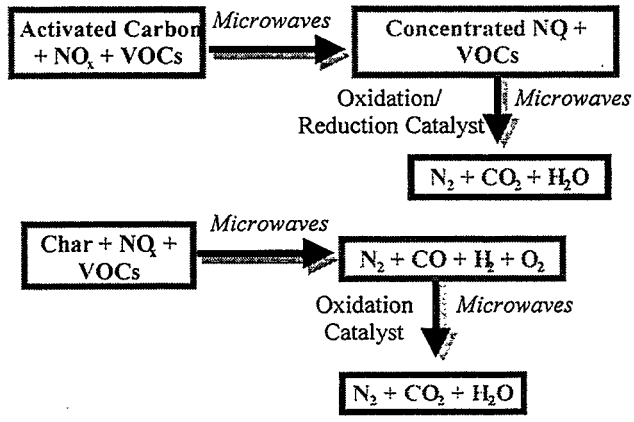
Figure 1



The CHA Corporation has been employing 2450 MHz microwave energy to drive various chemical reactions for several years[3,4]. We have found that NO_x and VOCs can be adsorbed by carbon and subsequently decomposed via chemical reduction by introd-ucing microwaves. Reaction temperatures are surprisingly low starting from 200 to 300 degrees Fahrenheit. We have also looked at various ways to utilize these reactions to abate NO_x and VOCs in diesel engine exhaust gases. The following group of reactions, shown in Figure 2, depict two schemes we have investigated. In the top set of reactions, the NO_x and VOCs are adsorbed on commercial activated carbon and the carbon is regenerated with microwave energy. In the second scheme the NO_x and VOCs are adsorbed on a less expensive carbon adsorbent labeled as char.

Figure 2

Interaction of Microwaves and Pollutants Adsorbed on Carbon



With the activated carbon scheme, the adsorbed pollutants are when microwave energy is introduced. The concentrated stream of NO_x and VOCs leaving the carbon is passed through an oxidation/re-duction catalyst bed where the pollutants are converted to harm-less gases. In the char scheme, a quite different sequence of reactions occur. In the presence of microwave energy, the adsorbed NO_x is reduced by the char to nitrogen and carbon monoxide. The VOCs are also reduced to hydrogen and carbon monoxide. Which gases are subsequently oxidized to carbon dioxide and water in an oxidation catalyst bed or monolith.

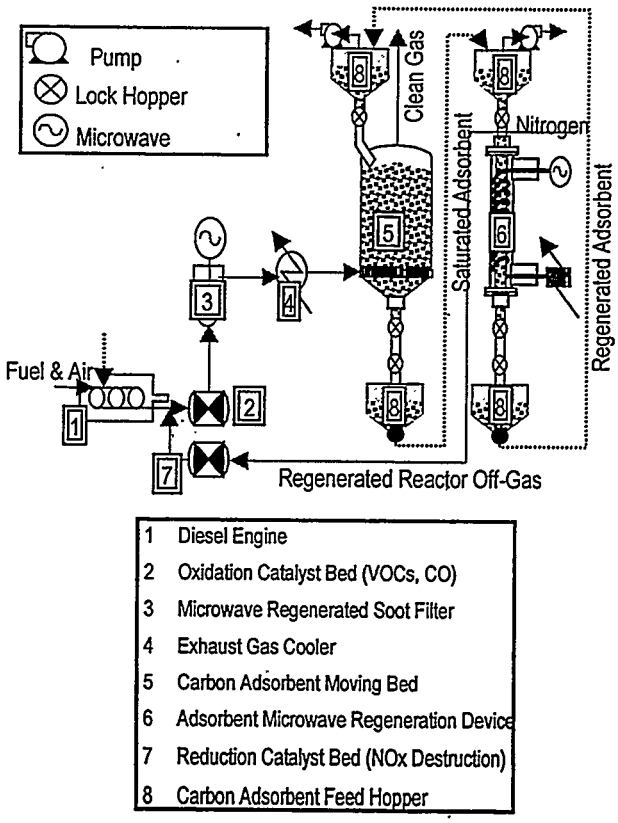
The process flow diagram for both schemes is shown in Figure 3.

Raw diesel exhaust gas is passed through a soot filter (3), heat exchanger (4), and carbon contactor (5), before release to the atmosphere. Activated carbon or char is an excellent adsorber for NO_x and VOCs. A NO_x capacity as high as 5.5% by weight can be achieved at gas temperatures below 175 degrees F. The NO_x adsorption capacity dominates saturation time since VOC concentrations are in the range of 150 to 400 ppm and NO_x concentrations are in the range of 500 to 2000 ppm. The carbon's adsorption ability is temperature dependent. No NO_x adsorption occurs above 300 degrees F and significant NO_x adsorption is not seen until the gas is cooled below 200 degrees F. Equally, the dew point of diesel exhaust is about 125 degrees F. This places the operating temperature range at 125 to 200 degrees F. Figure 4 shows NO_x breakthrough time as a func-tion of temperature for activated carbon. Since the gas leaving the exhaust manifold is about 1,000 degrees F., the gas must be cooled before NO_x can be removed via carbon adsorption. The air cooled heat exchanger (4), which has small passages to accommodate good heat removal performance, is vulnerable to soot plugging. The purpose of the soot filter (3) is to prevent plugging in the heat exchanger.

In the contactor (5) a 1.5 to 2 second residence time is needed to remove all of the NO_x from the exhaust gas. The speed of this NO_x removal step is dependent on a reaction that occurs in the carbon bed. The NO_x leaving the diesel engine is comprised primarily of NO, nitric oxide. Nitric oxide will not adsorb onto carbon in the process operating temperature range. However, in the presence of oxygen and carbon, nitric oxide converts to NO_2, nitrogen dioxide. Nitrogen dioxide is readily adsorbed by carbon. The long residence time is needed to facilitate the conversion of NO to NO_2. Interestingly, when activated carbon desorbs NO_x, the desorbed species is primarily NO.

Figure 3

CHA Corporation Diesel Exhaust Treatment Process Flow Diagram

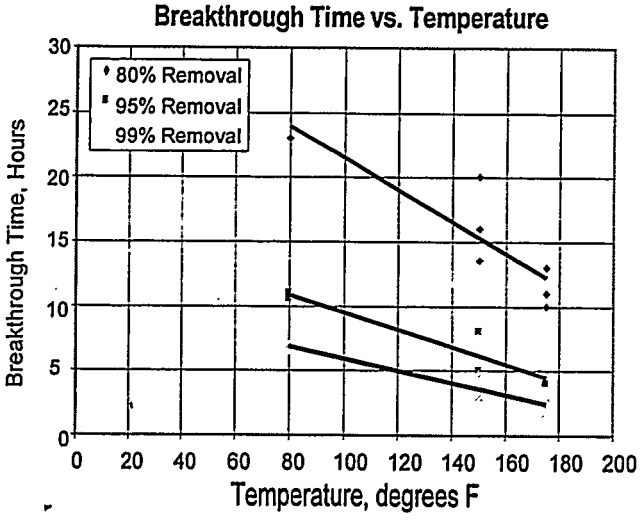


A moving bed contactor has been incorporated into the process to avoid NO_x breakthrough. Carbon is gravity fed through the bed and periodically lifted to the reactor feed hopper through a pneumatic lift system. In a similar fashion, spent carbon is gravity fed through the microwave reactor and pneumatically conveyed to the contactor feed hopper.

To size the prototype contactor and microwave regeneration reactor, we conducted a number of moving bed experiments using a 4.75 inch round contactor.

Diesel exhaust gas was passed through various carbon bed heights at various carbon feed rates and NO_x removal efficiency was determined for each configuration. Figure 5 shows a series of experimental results from this.

Figure 4

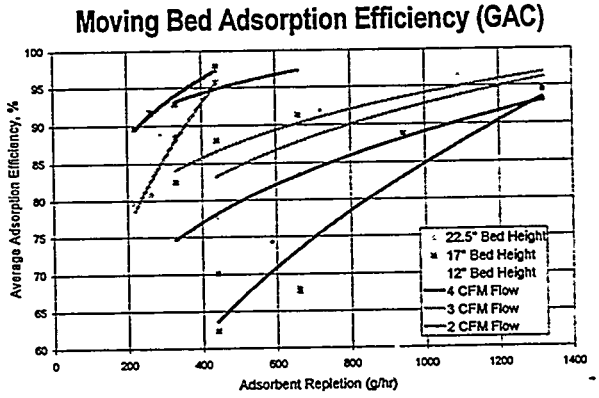


The contactor can be small in size and the carbon moved through at a high feed rate, or the contactor can be larger in size with a smaller carbon throughput rate. To continuously remove more than 90% of NO_x from a 100 SCFM gas stream carrying 1000 ppm NO_x, a square shaped contactor 30 inches in diameter is needed. A 22 inch tall carbon bed is needed to facilitate the NO to NO2 conversion and NO2 adsorption. Shorter beds (lower gas residence times) can be used if lower NO_x removal efficiency can be tolerated. For this size contactor, an activated carbon throughput of 15 pounds per hour is needed.

The microwave regeneration reactor was designed to match the 15 pounds of carbon per hour throughput of the contactor. Microwaves are applied through a 2.5 inch diameter 20 inch high helix wrapped around a 2.375 inch diameter quartz reactor tube at four wraps per foot. The quartz tube is held vertical in the reactor and the carbon is continuously gravity fed through the reactor tube. Microwave power level is constant at 1200 Watts. Carbon residence time in the reactor is 5.5 minutes. Nitrogen gas is purged through the reactor from top to bottom at 5 SCFH. The purge gas removes steam, desorbed pollutants and reaction products from the reactor and directs them to catalytic converter (7) in Figure 3, where desorbed pollutants are reduced. Microwave energy is 2,450 MHz. The source of the microwaves is two magnetrons identical to the ones used in a home microwave oven.

To test the process, we designed and constructed a prototype-integrated system to accommodate a 100 SCFM diesel exhaust stream. The prototype was matched to our 58 hp diesel test engine which is coupled to an alternator and load bank. This system is currently being evaluated at our laboratories and is scheduled for demonstration in March 1998.

Figure 5

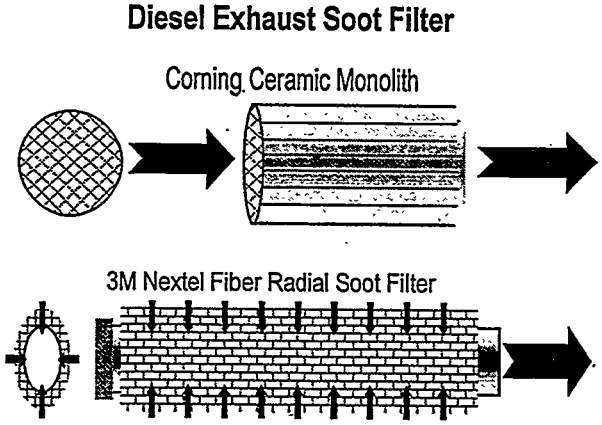


SOOT FILTER

Removing soot from the exhaust gases of a diesel has been a formidable task. Our need for a soot filter is limited to protecting the heat exchanger in the above mentioned process. As we

are interested in applying microwaves to various chemical processes, we were drawn to the idea of using microwaves to burn the soot from the surface of a filter material. Two materials were tested. One was a fiber based filter[5] manufactured by 3M and the other is a ceramic monolith[6] made by Corning, Inc. Figure 6 illustrates each of these soot filter materials. Samples of each soot filter were received from their respective manufacturers and suitable holders were prepared to pass diesel exhaust through the filter and subsequently apply microwave power to regenerate the filter.

Figure 6

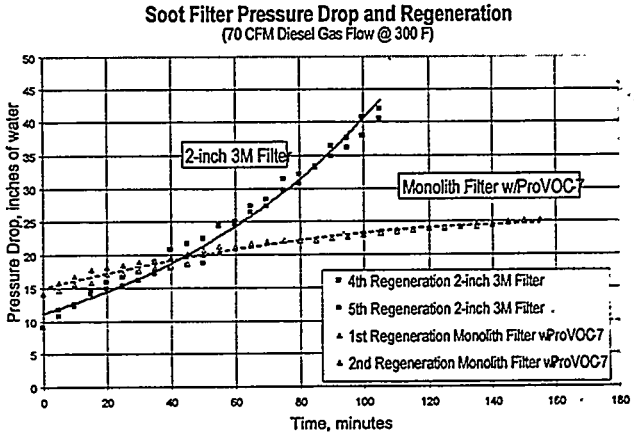


During regeneration, the filters were taken off line and a small flow of air was passed through the filter to supply oxygen. Microwave power levels used to regenerate the filters were in the range of 500 to 1500 Watts. Figure 7 shows the pressure buildup across each of these filters as a function of time. Two microwave regenerations are shown for each filter. Each ma-terial was successfully regenerated using microwaves. However, the filter material loaded with soot after two hours to a point where regeneration was needed. We are currently scaling up the size of these filters to increase the time between regenerations.

SUMMARY AND CONCLUSIONS

Diesel exhaust gases can be cleaned free of NO_x and VOCs by using activated carbon or char

Figure 7



that adsorb the pollutants. The carbon's ability to adsorb pollutants has been understood for some time. This work offers a method for regenerating the carbon adsorbent in a device operating in parallel to the absorber. The use of microwaves to desorb or destroy NO_x and VOCs on the surface of the carbon adsorbent is intriguing. The reaction proceeds quickly and reactor temperatures are low throughout carbon regeneration. Once tested and furhther developed, this process shows promise as a viable option for stationary diesel applications. The soot filter work shows that microwaves can be used to regenerate soot filters used to remove soot.

ACKNOWLEDGEMENT

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