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CATALYSIS OF CHARCOAL EMISSIONS

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ABSTRACT

This paper is about the development of a system to reduce the emissions of charcoal kilns with the catalyst from automotive catalytic converters. This device, called the charcoal kiln emissions oxidizer (CKEO), was designed and fabricated to meet the goal of a cost effective means to reduce the carbon monoxide and hydrocarbon emissions from charcoal kilns. Testing of the CKEO was never carried out as a test site could not be secured by this writing. The need for the CKEO, the theory behind it, and a procedure to test it are presented.

INTRODUCTION

The oxidation of charcoal kiln emissions through catalysis is a possible strategy for the charcoal kiln industry to follow in order to meet the stringent air pollution laws of the future. Currently the charcoal kiln industry is under no regulation. However, starting in 1995 \$5 for every ton of pollution will be charged to kiln operators by the Missouri Department of Natural Resources [1]. The trend toward higher regulation of this industry has its origin with the Clean Air Act.

Starting in 1978 charcoal kilns were given an indefinite exception to the Clean Air Act by the Missouri Conservation Department. The reason for this is that the Missouri Conservation Department concluded at the time that there was no reasonably available control (RAC) available for use by the kiln operators to reduce their exhaust emissions. The recent political climate has changed this attitude as well as the development of an RAC for the charcoal kilns in the State of Arkansas [2].

Emission regulation began in Arkansas after a State Supreme Court ruling in 1987. This regulation has been limited to visual inspection of particulates (smoke) released by the kiln. A system utilizing sawdust afterburners to reduce the smoke has been the resulting RAC. These afterburners are a containment vessel attached to the kiln's stack into which sawdust is automatically fed by augers when the exhaust temperature drops

below a predetermined level. This raises the temperature, and promotes a more complete combustion. Afterburners are not yet mandatory after four to five years of development, but they prove that there is an interest for some device to help regulate the pollutants created by charcoal kilns [3].

This investigation begins with a description of the characteristics of automotive catalytic converters. The investigation continues with a discussion of the design parameters of the CKEO, a description of the prototype CKEO built, and an outline of the testing procedure that was to be used.

AUTOMOTIVE CATALYTIC CONVERTERS

Dr. V.J. Flanigan of the University of Missouri--Rolla proposed the idea to reduce the emissions of charcoal kilns through the use of catalytic converters found on automobiles. Since the primary constituents of automobile and charcoal kiln emissions are similar, hydrocarbons (HC) and carbon monoxide (CO), an investigation of the catalyzing reaction upon charcoal kiln gasses is a reasonable undertaking.

Figure 1 shows the basic configuration common among most makes of catalytic converters.

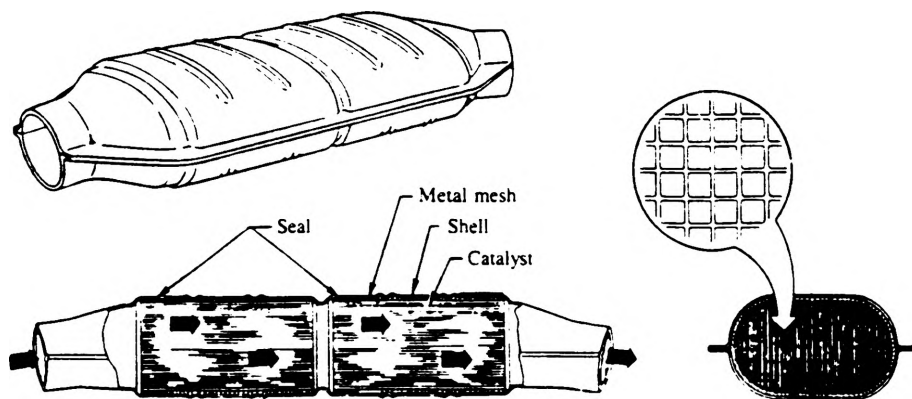


Figure 1. Catalytic Converter of Monolith Design [4].

The shell of the converter is made of stainless steel and contains the monolith. The converter catalyst is contained in the ceramic monolith that is made of alumina. The monolith is extruded into a square honeycomb configuration, and the squares are about one millimeter wide and tall. The catalyst is applied to the converter monolith with a washcoat of platinum (Pt), palladium (Pd), ruthenium (Ru), and rhodium (Rh). This washcoat deposits these noble metals upon the monolith surface with a particle size less than 50 nm and results in a huge surface area [5]. The inlet side monolith shown in the figure above is used for the oxidation of CO and HC into carbon dioxide and water, and its

catalyst is comprised mainly of Pt and Pd for this purpose. The exit side monolith in the converter is used to reduce oxides of nitrogen (NOx), and its catalyst is formulated to make use of Ru and Rh even though Pt and Pd are present too.

The important process under investigation is the oxidation of HC and CO gasses with atmospheric oxygen in the presence of a platinum catalyst. The oxidation of these reactants is rated by the conversion efficiency of the given system. The conversion efficiency is used to describe the effectiveness of the converter in reducing the mass flow rate of the undesirable CO and HC exhaust gasses by their oxidation. This is described in Equation 1 where “m” is the mass flow rate of a particular compound and “ η_{cat} ” is the conversion efficiency of the catalytic converter.

$$\eta_{cat} = 1 - (m_{out} / m_{in}) \quad (1)$$

The variables that affect the conversion efficiency of a given system are well known and are condensed into four items below:

- o The higher the oxygen content in the exhaust stream, the higher the conversion efficiency.
- o The higher the temperature, the higher the conversion efficiency.
- o The higher the temperature, the shorter the life of the catalyst.
- o The higher the catalyst surface area, the higher the conversion efficiency.

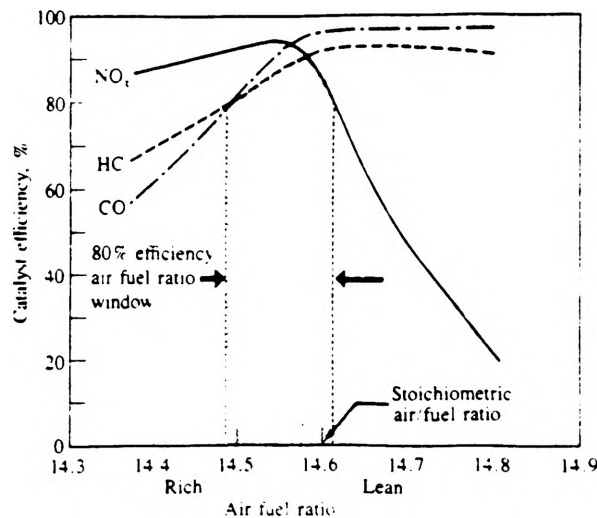


Figure 2. Conversion Efficiency for NO, CO, and HC as a Function of Exhaust Gas Air/Fuel Ratio [5].

The most important variable to control in automotive applications of catalytic converters is the oxygen content in the exhaust stream. There is a tight window of an engine's air/fuel ratio for catalytic converters to operate within as seen in Figure 2, but this investigation is only interested in the effects upon CO and HC so the restriction on high oxygen exhaust content imposed by NO_x does not apply. The figure shows that it is important for there to be an excess of oxygen for efficient oxidation of CO and HC to occur. Once sufficient amounts of oxygen are present, the next major variable to control for efficient oxidation is temperature.

The temperature has a great effect upon conversion efficiency. The temperature must be high enough for the reactants to oxidize in the presence of the catalyst. For automotive applications this temperature is referred to as the lightoff temperature. This is the temperature at which the catalyst operates at 50% conversion efficiency with an excess of oxygen. Below this temperature the catalyst is considered ineffective for automotive purposes. The lightoff temperatures for both HC and CO are in the range between 250 and 300 degrees Celsius. A desirable operating temperature would be one that produces at least 95% conversion efficiency temperature for the gasses involved. These temperatures are 425 degrees Celsius for CO and 310 degrees Celsius for HC [4]. These temperature effects are clearly seen in Figure 3.

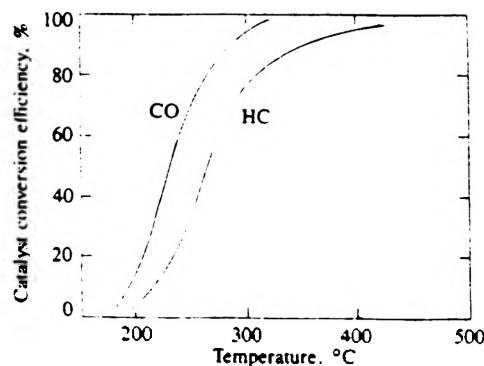


Figure 3. Conversion Efficiency for CO and HC as a Function of Temperature for Typical Oxidizing Catalytic Converter [4].

There is an upper limit imposed upon the operating temperature for noble metal catalysts due to the effects of sintering. When the noble metals are between 500 and 900 degrees Celsius, their sintering temperature range, their atoms begin to clump together. This lowers the exposed surface area of the catalyst as the particle sizes can increase to over 100 nm. This reduces the conversion efficiency of the catalyst if allowed to continue for even short periods of time [5].

Surface area of the catalyst can be chosen for automobiles through

a rule of thumb given by Kummer that says that the ceramic monolith's volume should be about half the displacement volume of an engine [5]. This should then provide adequate surface area for sufficient conversion of pollutants at the engine's operating speed. This ability to estimate the size of converter required for a given magnitude of exhaust flow rate is an important consideration in the design of the CKEO.

DESIGN PARAMETERS OF THE CKEO

The design of the CKEO involves the following parameters:

- o The volume of the catalytic converter must be sufficient to handle the exhaust volume produced by a charcoal kiln. This is estimated by the method suggested by Kummer.
- o There should be sufficient amounts of oxygen in order to produce at least 50% conversion efficiency at the operation temperature.
- o The operating temperature should be under the sintering temperature of 500 degrees Celsius and above the minimum lightoff temperature of 250 degrees Celsius.
- o The CKEO should not significantly increase the backpressure of the kiln exhaust.
- o The system should be as inexpensive as possible.

Since actual field testing was prevented and any reports concerning the operating parameters of charcoal kilns were not found, there are still questions concerning flow rate of kiln exhaust, oxygen content of kiln exhaust, temperature of kiln exhaust, and the effects of backpressure that the CKEO would have on the exhaust flow. A prototype CKEO was constructed for the purpose of field testing at a charcoal kiln facility with "best guesses" concerning the unknown parameters, and a testing procedure was outlined.

CONSTRUCTION OF THE CKEO

An unscaled sketch of the two pieces of the CKEO is shown in Figure 4. The construction considerations amounted to modifications to the converters and how to mount them on a charcoal kiln stack.

All monoliths were from used three-way 260 cubic-inch displacement catalytic converters from General Motors trucks equipped with 4.3 liter V-6 engines. This was estimated to provide an adequate catalyst surface area for the volume flow rate from a charcoal kiln. A catalytic converter that has been sawed in half between the monoliths and has

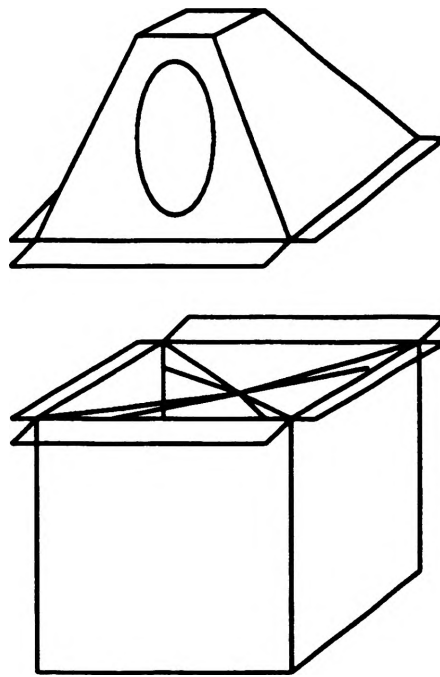


Figure 4. Sketch of CKEO.

the nozzle ends sawed off, to minimize flow restriction, is welded to each of the trapezoidal faces where the ellipse is shown in Figure 4. This configuration uses four monoliths from sectioned converters to shorten the length of the passage through the monolith which is essentially a laminar flow element. Calculations show that the use of four monoliths results in an opening that is 1.7 times larger than the six-inch diameter charcoal kiln exhaust stack for which the CKEO was designed, and should result in a fairly low flow restriction.

The top portion is made of stainless steel so that the catalytic converters will attach with a good weld, and it takes on a pyramidal shape in order to minimize the exposed surface area. This minimizes the heat transfer out of the exhaust gas and maintains as high a temperature as possible to maximize the conversion efficiency. Future testing may show that insulation around the top portion may increase the conversion efficiency further. The lower portion is welded together with cheaper non-stainless steel to keep the cost down. The cross bars on the lower section provide a structural stop for the charcoal kiln exhaust stack. A seven inch circular hole (not shown) on the bottom of the lower portion is where the exhaust stack enters the CKEO. The flanges on the top and bottom portions are drilled for bolts to attach the two sections. Openings between joined sections and the stack are to be sealed with fiberglass insulation prior to testing.

TESTING PROCEDURE

The following procedure was never implemented due to the fact that permission was not granted by a local charcoal kiln operator to perform testing. However, the following procedure was developed in anticipation of testing the CKEO at a charcoal kiln.

- o Develop a characteristic profile of the exhaust composition and temperatures during operation of a kiln. Since the cycle time of a charcoal kiln is about five to seven days, the profile should include at least two to three samples per day of the cycle in order to determine the oxygen levels in the exhaust and the maximum and minimum temperatures experienced. This will help indicate if some form of oxygen injection or addition of fuel to increase the temperature is required to maintain adequate conversion efficiency.
- o The CKEO that was built consisted of four types of converter monoliths. In this way the test could determine the effect of aging and different catalyst formulation. Three monoliths were Pt and Pd oxidizing catalysts from vehicles with the following mileages 20,000, 88,000, and an unknown number of miles. The fourth monolith was the NO_x reducing catalyst from the converter with 20,000 miles to determine how the different catalyst formulations would behave.
- o Temperatures were to be taken with type K thermocouples, and gas samples analyzed by the gas chromatography method. Temperatures and gas samples were to be taken from the stack from samples tubes welded on top of the CKEO. Also, temperature readings and gas samples were to be taken from the exiting stream of the monoliths, and surface temperatures along the side of the converters to determine if insulation would be useful in increasing conversion efficiency.

CLOSING REMARKS

It should be mentioned that even though testing of the CKEO was not accomplished, the background information concerning catalytic converters reinforces the feasibility of this concept rather than diminishing the practicality of the CKEO. Once the regulatory fines of the future are implemented on charcoal kiln operators, they will probably become more interested in a method to reduce the amount of pollutants released by the kilns and subsequently their fines.

ACKNOWLEDGEMENTS

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