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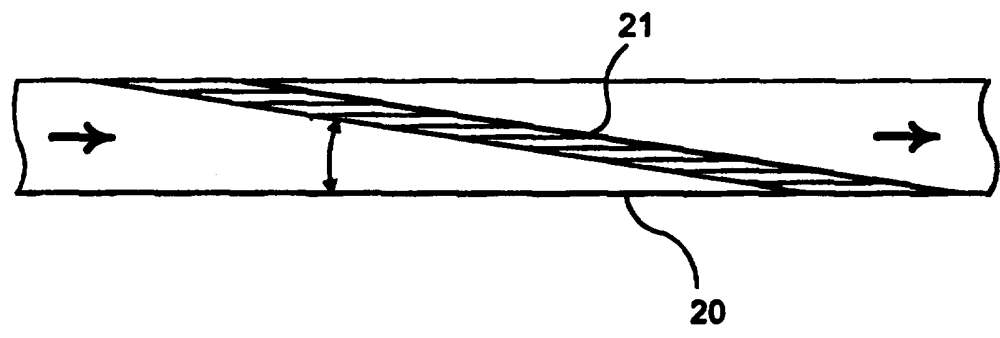
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(54) Title: CATALYTIC METHOD



(57) Abstract

A catalytic converter for control of exhaust gas pollutants comprising an oblique mounted monolith catalyst (11).

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CATALYTIC METHOD**BACKGROUND OF THE INVENTION****Field of the Invention**

5 This invention relates to improved catalytic
converters for control of automotive emissions. In
one specific aspect this invention relates to fast
light-off, low pressure drop catalytic converters.
It also relates to converters to achieve very high
10 conversions.

Brief Description of the Prior Art

Automotive emissions are still a major
environmental problem in spite of the major advances
brought about by the use of catalytic converters.
15 One factor limiting the performance of catalytic
converters is that pollution is not controlled during
the thirty or more seconds required to bring the
converter catalyst to its operating temperature. In
present converters, warm-up is dependent on heating
20 of the catalyst by hot engine exhaust gases.
Although electrical heating could be utilized to
preheat the catalyst prior to engine operation, the
power and the time delay required with present
catalyst structures, ceramic or metal, have been
25 deemed unacceptable.

Subsequent to catalyst light-off, surface
reactions on conventional monolithic catalysts such
as are used in catalytic converters are mass
transfer limited. Thus, the catalyst mass required
30 for a given conversion level is much higher than if
no mass transfer limitation existed at the given
operating conditions. The high catalyst mass
required for the required conversion level results in
the relatively long heat-up times experienced, even
35 with electrical heating.

A further problem is that catalyst diameter is much greater than the diameter of the inlet exhaust pipe. Consequently, exhaust gases tend to flow preferentially through the inner channels of the monoliths used, even with a diffuser inlet to spread out the flow.

The need to reduce catalyst warm-up time of the conventional ceramic monolith automotive catalysts to reduce emissions during the warm-up period has led to increased interest in metal monolith catalysts. However, merely substituting metal for ceramic in a conventional monolith structure yields catalysts which still have much too high a thermal mass. The short channel length catalysts of prior U.S. patent #5,051,241 offer the low thermal mass and high conversion efficiencies required. For automotive applications, packaging of such catalysts into a rugged assembly tolerant of flow pulsations is required to meet the new 100,000 mile durability requirements.

The present invention not only makes possible improved catalyst utilization of rugged fast light-off catalytic converters for automotive engine exhaust control which utilize the short channel length catalysts such as those of the above cited patent and the co-pending application filed February 23, 1994 (attorney docket #2885-23), but of conventional catalytic monoliths.

SUMMARY OF THE INVENTION

30

Definition of Terms

In the present invention the terms "monolith" and "monolith catalyst" refer not only to conventional monolithic structures and catalysts such as employed in conventional catalytic converters but also to short channel length structures of

enhanced mass transfer efficiency such as woven screens.

In the present invention the term "minilith" refers to monolith elements having flow channels of less than three millimeters in length and more than
5 forty channels per square centimeter.

For the purposes of this invention, the term "catalyst brick" refers to an assembly of minilith catalyst elements having channel flow passages less
10 than three millimeters in length and having more than forty channels per square centimeter and spaced apart by monolith elements of larger channel size.

The terms "carbonaceous compound" and "hydrocarbon" as used in the present invention refer
15 to organic compounds and to gas streams containing fuel values in the form of compounds such as carbon monoxide, organic compounds or partial oxidation products of carbon containing compounds.

The term "light-off" refers to the temperature
20 at which a catalyst achieves about fifty percent of the conversion achieved at the normal operating temperature.

The Invention

It has now been found that mounting a catalyst
25 brick or a monolith catalyst in an oblique position relative to the direction of exhaust gas inlet flow not only allows improved distribution through the catalyst flow channels, but also increased frontal catalyst area. Both result in a reduction in
30 pressure drop for a given conversion efficiency.

The increased catalytic frontal area achivable
with converters of the present invention allows a greater catalyst volume for a specified pressure drop, or alternatively use of much higher channel density,
35 to achieve a greater conversion efficiency in

pressure drop limited applications. The catalysts of prior U.S. patent #5,051,241 and the aforementioned application filed on February 23, 1994, incorporated herein by reference thereto, are especially
5 advantageous in the present invention.

Low pressure drop, rugged, high conversion efficiency and fast thermal response catalytic converters of the present invention using "catalyst bricks" make possible as much as a ten fold or more
10 reduction in catalyst mass as compared to that required to achieve the same conversion in mass transfer limited reactions of hydrocarbons using conventional monoliths catalysts and therefor are preferred catalysts for use in the present
15 invention.. As noted in the above referenced patent, it has been found that the specific mass transfer rate increases as the ratio of channel length to channel diameter of a monolith catalyst is reduced below about five to one or more preferably below
20 about two to one and especially below about one to one. Mass transfer of reactants to the surface becomes sensitive to the inlet flow rate rather than being significantly limited by the diffusion rate through a thick laminar flow boundary layer as in
25 conventional monolith catalysts, whether ceramic or metal. In conventional automotive monolith catalysts, the amount of pollutants oxidized is essentially independent of exhaust gas flow rate and thus percent conversion decreases with increase in
30 flow rate. In contrast, in the minilith catalyst assemblies, the amount of reactants oxidized typically increases with increase in flow rate. Thus if the inlet flow velocity is high enough, the reaction rate can even approach the intrinsic kinetic
35 reaction rate at the given catalyst temperature

without imposing an intolerable pressure drop. This means that it is practical to design automotive catalytic convertes for much higher conversion levels than is feasible with conventional catalytic
5 converters. Conversion levels of 99.9% or even higher are achievable in an automotive converter smaller in size than a lower conversion level conventional catalytic converter. Even conversion levels high enough for abatement of toxic industrial
10 fumes are achievable in compact reactors.

With the short flow paths, spaced apart catalysts, pressure drop is low permitting the use of much smaller channel diameters for a given pressure drop, further reducing catalyst mass required. The
15 rigid structure of catalysts bricks used in the present invention allows placement of a converter close to engine exhaust ports for more rapid heatup on starting an engine at low ambient temperatures. It has also been found that channel walls as thin as
20 0.1 mm or even less than 0.03 mm are practical with small channel diameters thus permitting high open areas even with such small channel diameters. Thus, as many as several thousand flow channels per square centimeter or even more are feasible without
25 reducing open area in the direction of flow below sixty percent. Open areas greater than 65, 70 or even 80 percent are feasible even with high channel density miniliths.

Inasmuch as heat transfer and mass transfer are
30 functionally related, an increase in mass transfer results in a corresponding increase in heat transfer. Thus, not only is catalyst mass reduced by use of the minilith catalysts of this invention, but the rate at which an automotive exhaust catalyst

is heated by the hot engine exhaust is correspondingly enhanced.

The reduced catalyst mass together with the increased heat transfer rate enables a short channel
5 catalyst to reach operating temperature much sooner than would a conventional automotive catalyst. If placed sufficiently close to the engine exhaust manifold, a minilith catalyst element can even reach operating temperature in less than ten seconds
10 without the need for electrical heating. Many alloys are commercially available which are suitable for metal miniliths of the present invention including Haynes alloy 25, Inconel 600, and even certain stainless steels. With metal microliths, alloy
15 selection is often determined primarily by oxidation resistance at the maximum operating temperature required by the given application.

The low pressure drops possible with catalytic converters based on the present invention makes it
20 possible to utilize a large number of small diameter elements, even as many as two hundred in a one inch length, such that the converter diameter is not significantly larger than the engine exhaust pipe. This makes it much easier to place the converter
25 catalyst at the exit of or even in the engine exhaust manifold, resulting in even faster catalyst warm up without electrical heating, and allows use of screens of different composition to achieve both hydrocarbon and NOx control. In fume abatement applications, the
30 large number elements feasible means that it practical to achieve whatever conversion levels are needed, even as high as 99.999 or better.

Although this invention has been described primarily in terms of automotive emissions, catalytic
35 reactors based on the present invention offer

advantages in any catalytic conversion system where reactor diameter is advantageously minimized or where it is desirable to minimize catalyst bed depth.

BRIEF DESCRIPTION OF THE DRAWINGS

5 Figure 1 shows a cross-sectional side view of a conventional monolith catalyst mounted in a conduit at an oblique angle to the direction of flow.

 Figure 2 shows a cross sectional side view of a converter with an oblique mounted monolith having
10 flow paths parallel to the direction of the flow through the converter.

DETAILED DESCRIPTION OF THE INVENTION AND PREFERRED EMBODIMENTS

 The present invention is further described in
15 connection with the drawings. As shown in the sectional view of figure 1, in one preferred embodiment a monolith catalyst 11 is mounted in housing 10 at an oblique angle relative to the inlet gas flow direction. Advantageously, the catalyst is
20 mounted at an angle of between about ten and forty degrees to the direction of flow.

 In the sectional view of figure 2, monolith 21 has flow paths essentially parallel to the direction of converter flow in converter 20 so that fluid
25 passing through converter 21 enters the flow channels with minimal change in direction as shown by the flow direction arrow thus allowing monolith 21 to be mounted at an oblique angle, ϕ , as little as five degrees. This reduces pressure losses.
30 Advantageously, monolith 21 is an assembly of minilith catalysts, preferably in the form of a catalyst brick such as described in pending patent application case attorney draft 2885-23 filed on February 23, 1994.

It should be noted that unlike parallel mounted catalyst beds, oblique mounting promotes a uniform fluid approach velocity across the catalyst inlet face. This is because the flow cross sectional area
5 decreases in approximate proportion to the flow volume.

The catalysts of the present invention are readily made using known catalytic agents. The following examples describe means of making minilith
10 catalysts but are not to be construed as limiting. A minilith catalyst of the present invention is made by vacuum sputtering platinum onto a stainless steel screen which has been cleaned by heating in air to 750K. Typically the platinum coating may be thinner
15 than 100 angstroms but may be thicker for greater catalyst life. Advantageously, a similarly thin layer of ceria or alumina may be deposited prior to deposition of the platinum. Catalysts containing palladium, iridium, rhodium or other metals can be
20 similarly prepared. In many applications, a wire screen formed from a catalytic alloy, such as a platinum doped alloy, is a sufficiently active catalyst without additional coating. Although metal miniliths are preferred, ceramic miniliths can be
25 made such as by slicing of ceramic honeycomb extrudates prior to firing. Such ceramic honeycomb extrudates advantageously may contain an organic binder to facilitate production of thin slices. However, ceramic miniliths are most advantageously
30 in the form of fiber mats or screens composed of long fibers spun from any desired ceramic composition, preferably catalytic ceramics. As necessary for sufficient low temperature catalytic activity, ceramic and metal miniliths may be catalyzed using
35 various techniques well known in the art.

EXAMPLE I

A multi-element catalytic microlith automotive exhaust reactor having forty minilith catalyst elements of 250 flow channels per square centimeter is constructed using a five centimeter wide strip of 70% open area screening of platinum coated stainless steel wires having a diameter of 0.10 mm with each screen spaced apart by a downstream screen having four channels per square centimeter with platinum coated wires 0.25 mm in diameter with the assembly clamped between two heavier screens of 1.5 mm diameter wires having one channel per square centimeter to form a catalyst brick nominally thirty centimeters long which is mounted in a container at an oblique angle of about nine degrees to the gas flow. Installed at the exhaust manifold outlet of a four cylinder automotive engine, catalyst light-off is within ten seconds of engine starting and thus exhaust emissions are controlled during initial operation of the engine.

EXAMPLE II

A fume abatement reactor is constructed with a catalyst brick having 100 elements of screening with about thirty 0.050 mm wires of platinum coated nichrome per centimeter (nominally 900 flow channels per square centimeter). Each element is spaced apart from the preceding one by a screen having nine 0.10 mm wires per centimeter. The catalyst brick is mounted at an oblique angle of five degrees to the gas flow to minimize pressure drop with a minimum reactor diameter. Fumes containing 50 ppm by volume of benzene in air are preheated to 700 degrees Kelvin and passed through the microlith reactor. Better than 99.9 percent conversion of the benzene to carbon dioxide and water is achieved.

CLAIMS

1. A reactor for catalytic conversion of hydrocarbons carried in a fluid flow, which comprises;

1. a conduit of predetermined effective diameter and predetermined flow cross-sectional area for carrying a hydrocarbon in a fluid flow on an axial flow path; and

2. a catalyst body having a plurality of elements with

a) a thickness less than the effective diameter of the conduit, and

b) multiple flow channels through the catalyst body; said catalyst body mounted in the conduit at an oblique angle to the axial flow path; said channels having a length of less than 3 millimeters and a density of more than 40 channels per square centimeter.

2. The reactor of claim 1 wherein said catalyst body is a monolith.

3. The reactor of claim 1 wherein said catalyst body is an assembly of miniliths.

4. The reactor of claim 1 wherein said catalyst body is a catalyst brick.

5. The reactor of claim 1 wherein said catalyst body is mounted an an angle of from 5° to 40° to the axial flow.

6. The reactor of claim 1 wherein the catalyst flow channels are aligned in the direction of axial flow.

7. The reactor of claim 1 wherein said fluid is the exhaust of an internal combustion engine.

8. The reactor of claim 1 wherein the catalyst body is mounted with the flow channels through the catalyst body in the direction of the axial flow path.

9. In the method for catalytic reaction of an organic compound carried in a fluid along a fluid flow path, which comprises:

5 passing the organic compound in a fluid flow through a plurality of open flow-through channels, each with a length of less than 3 millimeters in a catalyst body;

the improvement which comprises positioning the catalyst body at an oblique angle to the flow path;

10 said catalyst body having a thickness in the direction of flow less than the effective diameter of the flow path;

said channels having a density of more than 40 channels per square centimeter.

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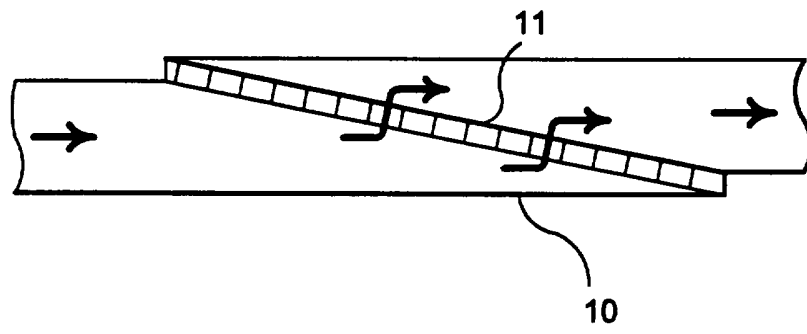


FIG. 1

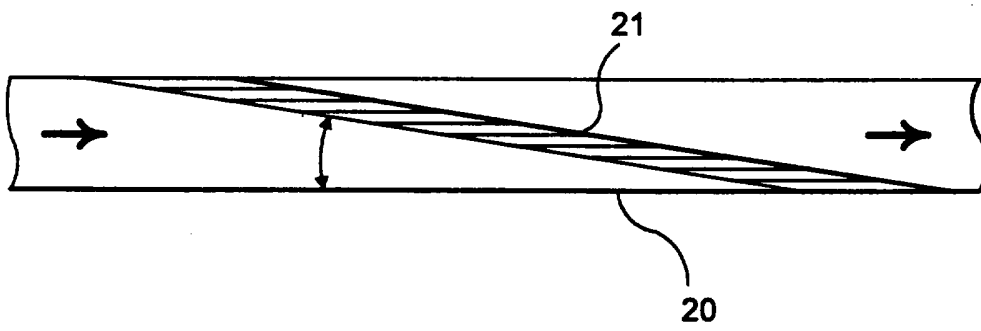


FIG. 2

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US95/10492

A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) :B01D 53/34; F01N 3/10
US CL :422/180, 177, 211, 222; 60/299; 502/527

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 422/171-175, 180, 177, 211, 222; 60/299; 502/439, 527; 55/dig. 30

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

NONE

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

NONE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US, A, 4,407,785 (PFEFFERLE) 04 October 1983, see entire document.	1-9
Y	US, A, 5,051,241 (PFEFFERLE) 24 September 1991, see entire document.	1-9
Y	US, A, 5,330,728 (FOSTER) 19 July 1994, see entire document.	1-9
Y	DE, A, 3,823,550 (BAYERISCHE MOTOREN WERKE) 18 January 1990, see abstract , Figure 2.	1-9

Further documents are listed in the continuation of Box C. See patent family annex.

* Special categories of cited documents:	*T	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
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