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(12) (19) (CA) **Demande-Application**

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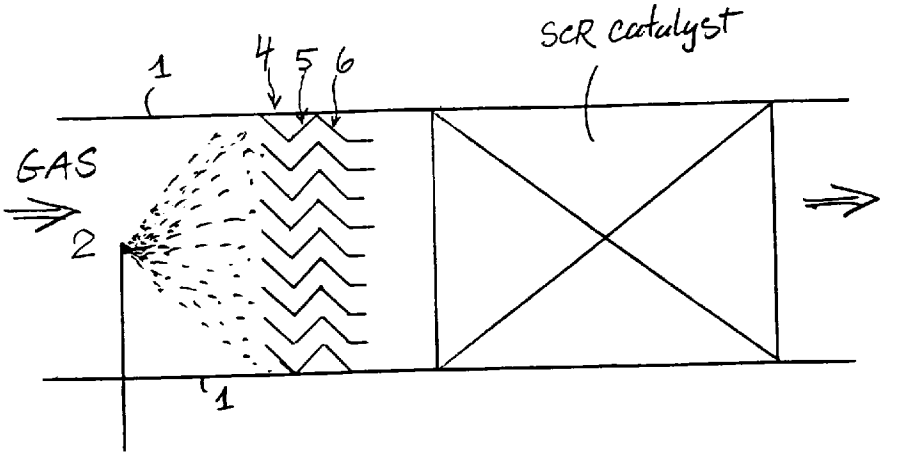
(54) **METHODE POUR LA REDUCTION SELECTIVE DES NO_X DANS LES GAZ D'ECHAPPEMENT**

(54) **PROCESS FOR SELECTIVE REDUCTION OF NO_X IN EXHAUST GAS**



(57) Méthode pour la réduction catalytique sélective des NOx dans les gaz d'échappement à des températures de 200-600°C, comprenant les étapes suivantes : a) injecter un brouillard de gouttelettes renfermant un réducteur et (ou) un précurseur de celui-ci, aptes à réduire les NOx, dans un conduit de gaz à travers un gicleur; b) faire passer les gaz d'échappement avec les gouttelettes entre les espaces délimités par plusieurs séries de plaques disposées parallèlement dans le conduit, une première série de plaques étant positionnée selon un angle de 45° environ par rapport aux parois du conduit de gaz, une seconde série de plaques formant un angle de

(57) Process for selective catalytic reduction of NOx in exhaust gases at temperatures of 200-600°C comprising the steps of: (a) injecting a spray of droplets containing a reductant and/or a precursor thereof being able to reduce NOx into a duct through a atomizing nozzle; (b) passing exhaust gas loaded with droplets through spaces between plates in a number of stacks of parallel plates in gas duct, wherein the plates in a first stack are positioned with an angle of about 45° relatively to walls of the gas duct, the plates of a second stack are positioned with an angle of about 90° relatively to the plates of the first stack and 45° relatively to the walls of the duct, a third stack with

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90° environ par rapport à la première série de plaques et 45° environ par rapport aux parois du conduit, enfin une troisième série de plaques étant parallèle à la première série de plaques, le tout forçant les gaz à passer à travers les trois séries de plaques en zigzaguant; c) après la dernière série de plaques, on fait passer les gaz à travers une couche de catalyseur monolithique pour la réduction sélective des NOx par les réducteurs via des canaux parallèles aux parois du conduit.

the plates being parallel to the plates of the first stack, whereby the gas is forced to pass through the stacks in zig-zag flow pattern; (c) after a last stack, passing the gas through a layer of monolithic catalyst for selective reduction of NOx by the reductant with channels, parallel to the walls of the duct.

ABSTRACT

Process for selective catalytic reduction of NOx in exhaust gases at temperatures of 200-600°C comprising the steps of:

(a) injecting a spray of droplets containing a reductant and/or a precursor thereof being able to reduce NOx into a duct through a atomizing nozzle;

(b) passing exhaust gas loaded with droplets through spaces between plates in a number of stacks of parallel plates in gas duct, wherein the plates in a first stack are positioned with an angle of about 45° relatively to walls of the gas duct, the plates of a second stack are positioned with an angle of about 90° relatively to the plates of the first stack and 45° relatively to the walls of the duct, a third stack with the plates being parallel to the plates of the first stack, whereby the gas is forced to pass through the stacks in zig-zag flow pattern;

(c) after a last stack, passing the gas through a layer of monolithic catalyst for selective reduction of NOx by the reductant with channels parallel to the walls of the duct.

(a) TITLE OF THE INVENTION

Process for Selective Reduction of NO_x in Exhaust Gases

(b) TECHNICAL FIELD TO WHICH THE INVENTION BELONGS

This invention relates to a process for the selective reduction NO_x which may be typically contained in exhaust gases from diesel engines by NH_3 that is generated by the decomposition of e.g., urea, solutions of urea in water or solutions of NH_3 in water, which is used for the selective reduction of NO_x by NH_3 in the gas phase upstream of the so-called SCR catalyst.

(c) BACKGROUND ART

The compound used as reductant is injected in liquid form as a spray, generated in a spray nozzle, into the exhaust gas stream having a temperature of typically 300-400°C that is required for the catalytic NOx reduction process. Droplets in the spray must, however, be evaporated and urea decomposed into NH_3, CO_2 and H_2O before the NH_3 can be utilized in the SCR catalyst for the NOx-reduction process. Evaporation and decomposition of droplets are strongly endothermal. The rate of evaporation and decomposition (gasification) of droplets is limited by the rate at which heat is transferred from the hot gas to the surface of the droplets through the gas film surrounding the droplets. This means that the time **t** for gasifying a droplet (the residence time) can be estimated by the equation:

$$t = K*d^{-2}*(T-Tmin.) \text{ seconds}$$

where **d** is the (initial) diameter of the droplet and **T** the temperature of the gas, Tmin. the minimum temperature required for decomposition of the urea at an appropriate rate (i.e. about 180°C), while **K** is a constant that is nearly independent on the gas velocity.

Thus, the residence time of the droplets increases strongly with the size of the droplets.

The distance between the injection nozzle and the SCR catalyst required for achieving complete gasification of the droplets in the gas stream, furthermore, increases proportional to the gas velocity in the duct.

It is estimated from experiments and calculations that a residence time of 100 μm droplets of 35% solution of urea in exhaust gas at 300°C is in the order of 1 second which means that a distance of 20 m from the nozzle to the SCR catalyst is required for complete gasification of the droplets at 20 m/s gas velocity in the duct. If only 0.5 m distance from injection nozzle to the SCR catalyst is available (as in the case of diesel vehicles), the size of the droplets must, therefore, be decreased to less than 8 μm, which is very difficult, in order to achieve complete gasification of the droplets in the gas phase upstream of the SCR catalyst.

Most of the particles or droplets in the gas will pass unreacted through the SCR catalyst when the catalyst is monolithic with straight channels throughout the catalytic block.

(d) DESCRIPTION OF THE INVENTION

It has now been found that relatively large droplets of a reductant which is generated by a simple spray nozzle, can be relatively-quickly and substantially-completely gasifies if the droplets are caused to impinge on surfaces of a body that can easily be installed at the limited distance available between the nozzle and the SCR catalyst in e.g., a diesel engine.

Thus, the present invention, in a broad aspect, now provides a process for the selective reduction of NO_x in exhaust gases at temperatures of 200 to 600°C., comprising the steps of:

(a) injecting a spray of droplets containing a reductant and/or a precursor thereof being able to reduce NOx into a duct through a atomizing nozzle;

(b) passing exhaust gas loaded with droplets through spaces between plates in a number of stacks of parallel plates in gas duct, wherein the plates in a first stack are positioned with an angle of about 45° relatively to walls of the gas duct, the plates of a second stack are positioned with an angle of about 90° relatively to the plates of the first stack and 45° relatively to the walls of the duct, a third stack with the plates being parallel to the plates of the first stack, whereby the gas is forced to pass through the stacks in zig-zag flow pattern;

(c) after a last stack, passing the gas through a layer of monolithic catalyst for selective reduction of NOx by the reductant with channels parallel to the walls of the duct.

(e) DESCRIPTION OF THE FIGURES

In the accompanying drawings:

Fig. 1 is a schematic sketch depicting the principles of aspects of the present invention:

Fig. 2 is a schematic view of the trajectories of the droplets;

Fig. 3 is a schematic view showing the efficiency of impingement when the gas passes to the next stack; and

Fig. 4 shows an alternative means for providing channels for gas flow.

(f) AT LEAST ONE MODE FOR CARRYING OUT THE INVENTION

The body is shown in principles in Fig 1. It consists of a number of consecutive stacks each stack consisting of parallel plates that cause the gas to flow in a zig-zag pattern in the spaces between the plates from stack to stack whereby the droplets tend to impinge on the surface of the plates each time that the direction of flow is changed. When the droplets impinge, they splatter out on the surface on the surface whereby the liquid is heated up

at a much higher rate than in the form of droplets in the gas phase because the plates are heated much more efficiently by the gas for several reasons. Hence, the plates enlarge the surface of heat transmission between liquid and gas, and the gas film restriction for heat transfer between the stagnant plates and the gas is much higher than that of droplets moving passively along with the gas flow.

The tendency for droplets to impinge on the plates of a stack increases with increasing size of the droplets, with increasing gas velocity in the space between the plates, with increasing angle of deflection of the gas stream when passing from stack to stack with the length of the plates in the direction of flow and with decreasing distance between the plates in a stack. This is shown in Fig. 2 showing calculated trajectories of 20, 50 and 100 μm droplets in a gas passing through 3 stacks of plates arranged with a distance of 10 mm between the plates and 90° angle of deflection of the gas passing from stack to stack and a gas velocity of 8 m/s in the spaces between the stacks. It is seen that the 100 μm droplet impinge already in the first stack, 50 μm droplet have impinged after the third stack, while the 20 μm droplet passes through all three stacks.

The efficiency of impingement will increase when the position of the plates in one stack are displaced from the position of the plates in the next stack so that the spaces between the plates are cut in half, when the gas passes to the next stack as shown in Fig. 3.

The rate of gasification and, in particular, the minimum temperature of the gas required for obtaining complete gasification of the reductant can be increased and decreased, respectively, coating the surfaces of the plates, or at least the areas of impingement of the plates, by materials such as oxides of Al, Fe, Ti, Zr, Si, Zn and/or Mg, which catalyze the decomposition of urea.

Impingement and mixing of the gas can be combined by staggering the plates in principles in the same way as used in known types of stationary mixers. The impingement effect will, however, be decreased when combined with mixing of the gas because of the bypass of gas between the channels in stationary mixers, which at a given mixing effect decreases the deflection of the gas.

Instead of sharp edged channels formed by stacks of flat plates as shown in Fig. 1, the channels for gas flow can, alternatively, be formed as shown in Fig. 4 by piling wave-shaped plates with the waves perpendicular to the walls of the duct. The gas flow in the channels formed by the spaces between the wave-shaped plates will substantially be deflected in the same way as in sharp-edged channels formed by spaces between the above described array of flat plates shown in Fig 1.

CLAIMS

1. Process for selective catalytic reduction of NOx in exhaust gases at temperatures of 200-600°C comprising the steps of:

(a) injecting a spray of droplets containing a reductant and/or a precursor thereof being able to reduce NOx into a duct through a atomizing nozzle;

(b) passing exhaust gas loaded with droplets through spaces between plates in a number of stacks of parallel plates in gas duct, wherein the plates in a first stack are positioned with an angle of about 45° relatively to walls of the gas duct, the plates of a second stack are positioned with an angle of about 90° relatively to the plates of the first stack and 45° relatively to the walls of the duct, a third stack with the plates being parallel to the plates of the first stack, whereby the gas is forced to pass through the stacks in zig-zag flow pattern;

(c) after a last stack, passing the gas through a layer of monolithic catalyst for selective reduction of NOx by the reductant with channels parallel to the walls of the duct.

2. The process of claim 1, wherein substantially-all said droplets impinge on the walls of the plates in the stacks, the diameter of the droplets are larger than a critical diameter at which the droplets are caused to impinge on said walls whenever the direction of gas flow is deflected through passage in the stacks.

3. The process of claim 1 or claim 2, wherein said droplets

4. The process of claim 1, claim 2 or claim 3, wherein said plates are metallic.

5. The process of claim 4, wherein said metallic plates are formed of an acid-resistant stainless steel alloy.

6. The process of claim 1, claim 2 or claim 3, wherein said plates consist of ceramic material.

7. The process of claim 6, wherein said plates consist of cordierite.

8. The process of any one or more of claims 1 to 7, inclusive, wherein said plates are coated, at least on the surfaces which are exposed to said impinging droplets, by a 0.01 to 1 mm layer of at least one of the oxides of Al, Fe, Ti, Si, Zn or Mg.

9. The process of any one or more of claims 1 to 8, inclusive, wherein said reductant is a liquid mixture of urea and water with 30 to 100 weight % urea.

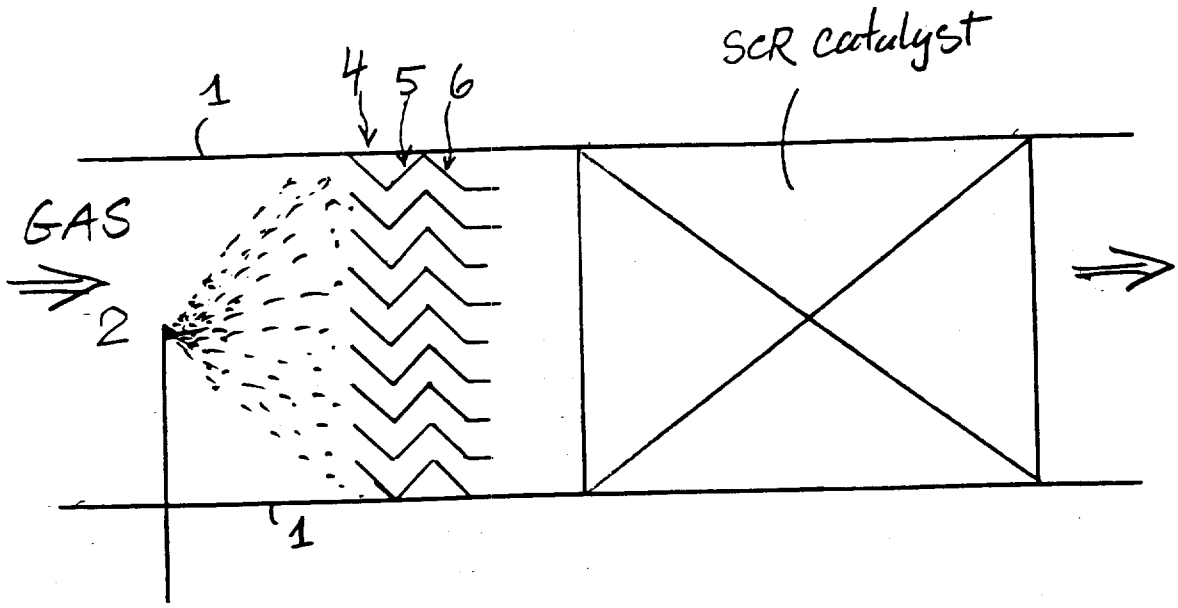

Fig 1

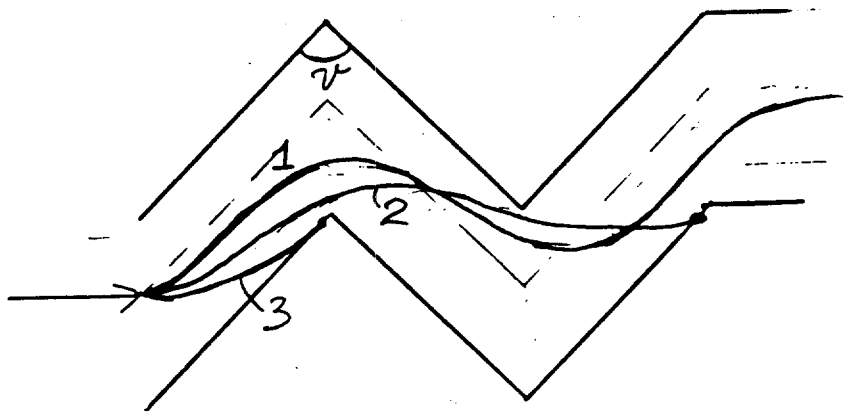

Fig 2. Trajectories of:
1 20 μm droplet
2 50 " "
3 100 " "

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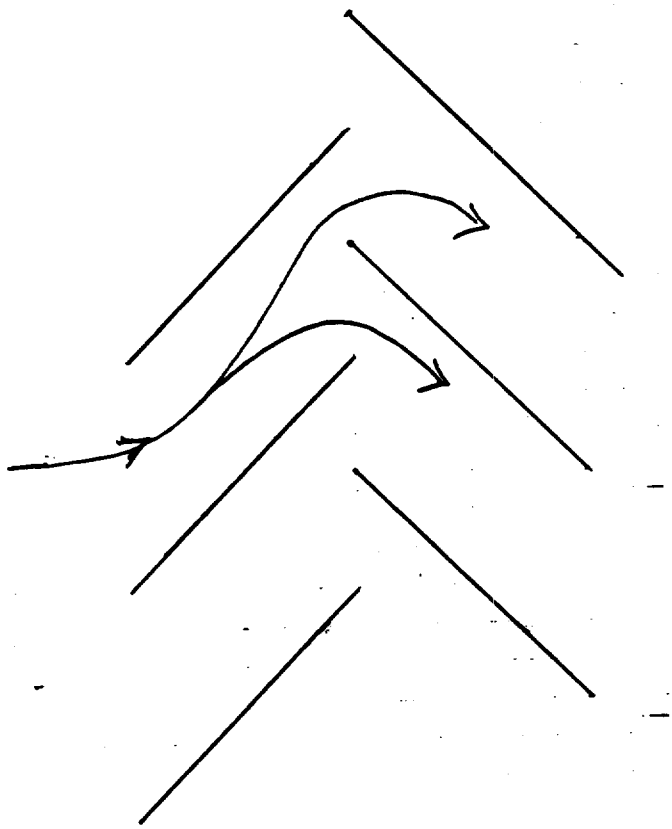

Fig 3

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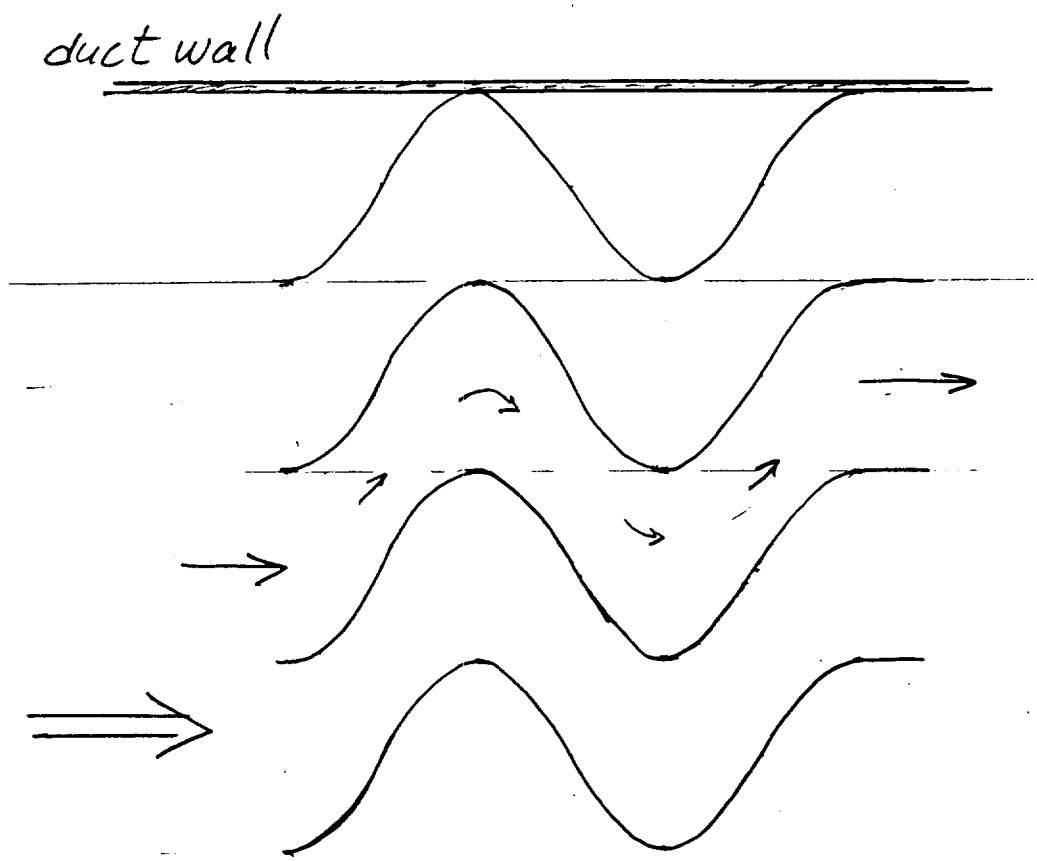

Fig 4.

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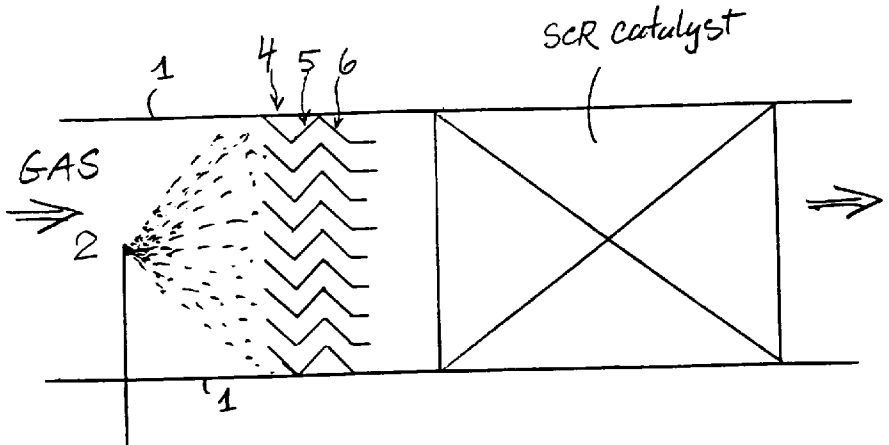
SCR catalyst
1
4
5
6
GAS
2
1