

METHOD FOR ADSORBING AND DECOMPOSING LEAN NITROGEN OXIDE
USING A MIXED METAL OXIDE CATALYST

Technical Field

5 The present invention relates to a method of
adsorbing and reducing nitrogen oxides using a mixed
metal oxide catalyst, and more particularly, to a method
of improving the efficiency of adsorption and desorption
of nitrogen oxides using a mixed metal oxide catalyst
10 prepared from a hydrotalcite precursor.

Background Art

 Nitrogen oxides (NO_x) are typical air pollutants
which cause smog, acid rain, photochemical smog by
15 atmospheric photochemical reactions and various
secondary pollutants (O₃, PAN, etc.) harmful to the human
body and contribute to the eutrophication of aquatic
ecosystems.

 Also, nitrogen oxides are currently regulated
20 emissions produced unavoidably from the combustion of
most fossil fuels. As methods for decomposing and
removing nitrogen oxides, selective catalytic reduction
(SCR) technology is used, which utilizes three-way
catalysts for gasoline engines, and NH₃ and hydrocarbons,
25 etc., as reducing agents.

 In such methods, the efficiency of decomposition
of nitrogen oxides decreases rapidly when the nitrogen
oxides coexist with other components such as oxygen. In
addition, the selective catalytic reduction method that
30 has been put into practical use has problems in that the
range of reaction temperature is narrow, the performance
of the catalyst is deteriorated due to aging, and set-up
and operating costs are high. Thus, research and
development have been continuously conducted in order to
35 solve these problems.

Oxygen (O_2) is present in exhaust gases in case that a sufficient amount of oxygen is supplied for complete combustion of fossil fuels. In this case, the remaining oxygen greatly increases the consumption of NOx reducing agents or greatly reduces the reduction ability thereof. Thus, there is required a method that adsorbs NOx and then desorbs NOx, thereby preventing the NOx reduction ability of the reducing agent from being deteriorated due to the presence of oxygen and effectively achieving the decomposition of NOx. According to this requirement, the research and development of NOx adsorbing materials have been conducted.

NOx adsorbing materials are based on alkaline earth metal oxides such as BaO. The adsorption capacity of these oxides for NO is significantly lower than that for NO_2 . For this reason, a catalyst for oxidizing and adsorbing NO needs to be used. As this catalyst, Pt that is an expensive transition metal is mainly used. However, the maximum adsorption capacity of these adsorbing materials is generally only 10~20 mg-NOx/g-cat at 350 °C, and significantly deteriorates as temperature increases or decreases. In addition, because these adsorbing materials are based on NO_2 -adsorbing components rather than NO-adsorbing components, their ability to decompose NOx in the desorption process is low, so that these adsorbing materials cannot act as effective adsorbing materials.

Patent document 1 discloses the reductive decomposition of nitrogen oxides. However, the patent document relates to the reductive decomposition of nitrogen oxides in case that no oxygen is present in exhaust gases. It is difficult to apply to the case in which oxygen is present.

Patent document 2 discloses a method of

decomposing nitrogen oxides using carbon monoxide (CO). However, the patent document has a shortcoming in that, when oxygen is present, CO corresponding to the equivalent of oxygen is additionally required.

5 In addition, patent document 3 adopts a method of supplying CO resulting from partial oxidation of CH₄ and discloses a method of decomposing N₂O and NO_x using an MMO catalyst. However, in order to decompose lean NO_x and N₂O, a method different from patent document 3 is
10 required.

 If oxygen is present in excess, it can be present in an amount much larger than that of NO_x, which is at most several thousand ppm or less, and thus the consumption of CO for removing this oxygen will be very
15 high. In this case, NO_x treatment costs will be increased due to the consumption of the reducing agent, it will be difficult to find a suitable method for supplying CO, and the amount of CO₂ discharged can also be problematic.

20 Patent Document 1: Korean Patent Registration No. 563903

 Patent Document 2: Korean Patent Registration No. 654885

 Patent Document 3: Korean Patent Registration No.
25 638835

Summary

According to one aspect, there is provided a method for decomposing nitrogen oxides using a mixed metal oxide catalyst, the method comprising the steps of:

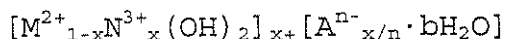
i) providing a catalyst layer comprising a mixed metal oxide catalyst prepared from a hydrotalcite-type or brucite-type compound precursor represented by the following formula 1, wherein the order of adsorption strength of adsorbants thereon is $(\text{NO}, \text{N}_2\text{O}, \text{NO}_2, \text{SO}_2) > \text{CO} > \text{O}_2$, a catalyst wherein a precious metal is impregnated or intercalated into the mixed metal oxide catalyst, or a catalyst wherein a co-catalyst is added and a non-precious metal is impregnated or intercalated into the mixed metal oxide catalyst;

ii) passing a waste gas containing oxygen and nitrogen oxides and having an oxygen concentration of 0.01% or more, through the catalyst layer, to selectively adsorb the nitrogen oxides on the catalyst layer;

iii) passing a reducing agent through the catalyst layer having the nitrogen oxides adsorbed thereon; and

iv) decomposing and desorbing the adsorbed nitrogen oxides by the reducing agent:

[Formula 1]



wherein M^{2+} is selected from the group consisting of Mg^{2+} , Ca^{2+} , Ni^{2+} , Zn^{2+} , Sr^{2+} , Ba^{2+} , Fe^{2+} , Cu^{2+} , Co^{2+} , Pb^{2+} and Mn^{2+} ; N^{3+} is selected from the group consisting of Al^{3+} , Mn^{3+} , Fe^{3+} , Co^{3+} , Ni^{3+} , Cr^{3+} , Ga^{3+} , B^{3+} , La^{3+} , Ce^{3+} and Rh^{3+} ; A^{n-} is selected from the group consisting of CO_3^{2-} , NO_3^- , SO_4^{2-} , Cl^- , OH^- , SiO_3^{2-} , MnO_4^{2-} , HPO_3^{2-} , MnO_4^{2-} , HGao_3^{2-} , HVO_4^{2-} , ClO_4^- and BO_3^{2-} ; x ranges from 0.0 to 0.5; and b is an integer ranging from 0 to 20.

Detailed Description of the Invention

Technical Problem

The present inventors have made extensive efforts to solve the above-described problems occurring in the prior art, and as a result, have found that the efficiency of decomposition of NO_x and N_2O can be greatly improved even in the presence of oxygen (O_2) by selectively adsorbing and separating nitrogen oxides including NO_x and N_2O from the coexisting oxygen using a

mixed metal oxide catalyst prepared from a hydrotalcite-type precursor, and then desorbing the adsorbed nitrogen oxides. Based on this finding, the present invention has been completed.

5 Therefore, it is an object of the present invention to provide a method which uses a mixed metal oxide catalyst, prepared from a hydrotalcite-type precursor, to efficiently perform the decomposition of NO_x and N₂O, which coexist with oxygen (O₂) known to be
10 very difficult to reduce and decompose.

Technical Solution

In order to achieve the above object, the present invention provides a method for decomposing lean
15 nitrogen oxides, comprising the steps of:

i) providing a catalyst layer comprising a mixed metal oxide catalyst prepared from a hydrotalcite-type compound precursor, a catalyst wherein a precious metal is impregnated or intercalated into the mixed metal
20 oxide catalyst, or a catalyst wherein a co-catalyst is added and a non-precious metal is impregnated or intercalated into the mixed metal oxide catalyst;

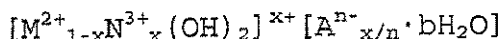
ii) passing a waste gas, containing oxygen and nitrogen oxides and having an oxygen concentration of
25 0.01% or more, through the catalyst layer, to selectively adsorb the nitrogen oxides on the catalyst layer;

iii) passing a reducing agent through the catalyst layer having the nitrogen oxides adsorbed thereon; and

30 iv) decomposing and desorbing the adsorbed nitrogen oxides by the reducing agent.

In one embodiment of the present invention, the hydrotalcite-type precursor is represented by the following formula 1:

35 [Formula 1]



wherein M^{2+} is selected from the group consisting of Mg^{2+} , Ca^{2+} , Ni^{2+} , Zn^{2+} , Sr^{2+} , Ba^{2+} , Fe^{2+} , Cu^{2+} , Co^{2+} , Pa^{2+} and Mn^{2+} ; N^{3+} is selected from the group consisting of Al^{3+} , Mn^{3+} , Fe^{3+} , Co^{3+} , Ni^{3+} , Cr^{3+} , Ga^{3+} , B^{3+} , La^{3+} , Ce^{3+} and Rh^{3+} ; A^{n-} is selected from the group consisting of CO_3^{2-} , NO_3^- , SO_4^{2-} , Cl^- , OH^- , SiO_3^{2-} , MnO_4^{2-} , HPO_3^{2-} , MnO_4^{2-} , $HGaO_3^{2-}$, HVO_4^{2-} , ClO_4^- and BO_3^{2-} ; x ranges from 0.0 to 0.5; and b is an integer ranging from 0 to 20.

10 In one embodiment of the present invention, the order of adsorption strength of adsorbants on the hydrotalcite-type or brucite-type precursor is $(NO, N_2O, NO_2, SO_2) > CO > O_2$.

In one embodiment of the present invention, SO_2 is desorbed from the catalyst by any one or more of CO , NO , NO_2 , O_2 and N_2 in case that SO_2 is adsorbed onto the catalyst such that a decrease in the activity of the catalyst by the accumulation of SO_2 does not occur.

20 In one embodiment of the present invention, the precious metal is selected from the group consisting of Mo , Ti , Pt , Au , Ag , Rh , Pa , La , Ir , V , Kr , Nd , Nb , Se , Sc , Ru , In , Y , Z , and mixtures thereof, and the amount of the precious metal in the intercalated catalyst is preferably 0.01 to 50 wt%.

25 In one embodiment of the present invention, the co-catalyst is selected from among alkali metals, and the non-precious metal is selected from the group consisting of Ni , Co , Fe , Mn , Al , Ce , Zr , Cu , Zn , Ba , Mg , Ca , Sr , and mixtures thereof. In addition, the content of the non-precious metal in the intercalated catalyst is 0.01 to 99.9 wt%.

35 In one embodiment of the present invention, the reducing agent further comprises a component selected from the group consisting of CO , H_2 , C_1-C_6 hydrocarbons, C_1-C_6 alcohols, and mixtures thereof.

In one embodiment of the present invention, the hourly space velocity of gas during passage through the catalyst layer is $1,000\text{h}^{-1}$ to $300,000\text{h}^{-1}$.

5 In one embodiment of the present invention, the reaction temperature is preferably 20 to $1,000\text{ }^{\circ}\text{C}$.

In one embodiment of the present invention, the waste gas preferably comprises 0.001-30 wt% of N_2O , 0.001-10 wt% of NO, 0.001-21 wt% of O_2 , 0.001-20 wt% of the reducing agent, and a balance of other contaminants
10 and inert carrier gases.

Advantageous Effects

According to the method of the present invention, NOx and N_2O in a waste gas, which have an oxygen
15 concentration of 0.01% or more and contain NOx and oxygen; or NOx, N_2O and oxygen, known to be very difficult to reduce and decompose, can be efficiently decomposed by adsorbing and separating NOx and N_2O from oxygen and then decomposing the adsorbed NOx and N_2O
20 using a reducing agent.

In addition, adsorbing materials mainly adsorb NO_2 , and thus an expensive catalyst such as Pt should be used to oxidize NO and an increased amount of a reducing agent should also be used for desorption of NO. On the
25 other hand, when the mixed metal oxide (MMO) catalyst as described in the present invention is used, it shows a higher adsorption capacity for NO, and thus is advantageous in terms of cost.

30 Best Mode for Carrying Out the Invention

Hereinafter, the present invention will be described in further detail.

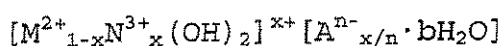
As described above, the present invention is characterized in that lean N_2O or NOx and N_2O are
35 adsorbed and separated from oxygen in the presence of a

mixed metal oxide catalyst prepared from a hydrotalcite-type or brucite-type and are decomposed and desorbed in an oxygen-free state.

Hydrotalcite compounds are generally based on anionic clays and are composed of hydrates of magnesium or aluminum, which are abundant on the earth. These hydrotalcite compounds can be synthesized at room temperature and atmospheric pressure. Until now, hydrotalcites have been used mainly as precursors of oxide catalysts, and supported or impregnated or intercalated hydrotalcite compounds have not yet been put into practical use.

The hydrotalcite precursor that is used in the present invention can be represented by the following formula 1:

[Formula 1]



In the hydrotalcite-type or brucite-type precursor, M^{2+} and N^{3+} are metal cations, respectively. Specifically, M^{2+} is a divalent metal cation selected from the group consisting of Mg^{2+} , Ca^{2+} , Ni^{2+} , Zn^{2+} , Sr^{2+} , Ba^{2+} , Fe^{2+} , Cu^{2+} , Co^{2+} , Pd^{2+} and Mn^{2+} , and N^{3+} is a trivalent metal cation selected from the group consisting of Rh^{3+} , Al^{3+} , Mn^{3+} , Fe^{3+} , Co^{3+} , Ni^{3+} , Cr^{3+} , Ga^{3+} , B^{3+} , La^{3+} and Ce^{3+} . The molar ratio of $M^{2+}:N^{3+}$ is 1:1 to 100:1, and preferably 1:1 to 5:1. In the hydrotalcite-type precursor, A^{n-} is an anionic compound having a charge of -1, -2 or -3 and is one or a combination of two or more selected from the group consisting of CO_3^{2-} , NO_3^- , SO_4^{2-} , Cl^- , OH^- , SiO_3^{2-} , MnO_4^{2-} , HPO_3^{2-} , MnO_4^{2-} , $HGaO_3^{2-}$, HVO_4^{2-} , ClO_4^- and BO_3^{2-} , x ranges from 0.0 to 0.5, and b is an integer ranging from 0 to 20.

The hydrotalcite-type precursor is calcined at a temperature of 100 to 1,000 °C to prepare a mixed metal oxide. In order to increase the catalytic activity of

the mixed metal oxide catalyst, at least one precious metal selected from among Mo, Ti, Pt, Au, Ag, Rh, Pd, La, Ir, V, Kr, Nd, Nb, Se, Sc, Ru, In, Y and Zr may optionally be impregnated or intercalated into the mixed metal oxide.

The amount of the precious metal in the intercalated catalyst is preferably 0.01 to 50 wt%.

In one embodiment of the present invention, the mixed metal oxide (MMO) catalyst that is used in the method of the present invention may be impregnated or intercalated with a non-precious metal. In general, a precious metal is expensive, but the use of a co-catalyst allows a non-precious metal such as Ni to substitute for a precious metal.

The co-catalyst that is used in the present invention may be an alkali metal. Preferably, it is selected from the group consisting of Na, K, Rb, and Cs. Moreover, the non-precious metal may be one or more selected from the group consisting of Ni, Co, Fe, Mn, Al, Ce, Zr, Cu, Zn, Ba, Mg, Ca, Sr, and mixtures thereof. The amount of the non-precious metal in the intercalated catalyst is preferably 0.01 to 99.9 wt%.

The present invention uses the adsorption capacity of the MMO catalyst, and the order of adsorption strength of adsorbants on the MMO catalyst is generally $(\text{NO}, \text{N}_2\text{O}, \text{NO}_2, \text{SO}_2) > \text{CO} > \text{O}_2$. Namely, the adsorption strength of NO, N₂O, NO₂ and SO₂ is higher than that of CO and O₂, but the order of adsorption strength between NO, N₂O, NO₂ and SO₂ can vary depending on, for example, the composition of the catalyst.

As described above, NO and N₂O are more easily adsorbed than oxygen. Thus, when an exhaust gas containing an excessive amount of oxygen passes through the catalyst layer for adsorption, NO and N₂O can be more adsorbed than oxygen. In other words, the majority of

oxygen passes through the catalyst layer while NO and N₂O are adsorbed on the catalyst and thus separated from oxygen. The separated NO and N₂O are then easily reduced by a reducing agent such as CO, and the reaction with
5 oxygen can be eliminated. Thus, the consumption of CO is low and the rate of desorption of NO and N₂O from the catalyst layer is fast.

The MMO catalyst generally shows a higher adsorption capacity for NO. SO₂ known as a catalyst
10 poison is strongly adsorbed compared to CO, O₂ and the like, but shows adsorption strength similar to NO, NO₂ and the like. However, according to the present invention, SO₂ adsorbed on the catalyst layer is desorbed by CO, NO, NO₂, O₂ and N₂, a decrease in the NOx
15 adsorption efficiency or activity of the catalyst, caused by the accumulation of SO₂, does not substantially occur.

In the inventive method of adsorbing and decomposing lean NOx, or NOx and N₂O using the mixed
20 metal oxide catalyst prepared from the hydrotalcite-type precursor, a waste gas that passes through the catalyst layer comprises 0 to 30 wt% of N₂O, 0.001 to 10 wt% of NO, 0.001 to 20 wt% of a reducing agent, 0.001 to 21 wt% of O₂, and the balance of other contaminants and inert
25 gases such as N₂.

If the amount of the reducing agent in the gas mixture is less than 0.001 wt%, it will not sufficiently reduce NO and N₂O into N₂ and O₂, and if it is more than
20 wt%, large amounts of unreacted reducing agent and reaction by-products will be produced, and thus another
30 process for removing them will be required.

In the present invention, the reducing agent may be one or more selected from the group consisting of CO, H₂, C₁-C₆ hydrocarbons, C₁-C₆ alcohols, and NH₃.

35 In the catalytic decomposition reaction of the

present invention, the gas hourly space velocity (GHSV) of the gas during passage through the catalyst layer is between $1,000 \text{ h}^{-1}$ and $300,000 \text{ h}^{-1}$, preferably between $30,000$ and $200,000 \text{ h}^{-1}$. If the GHSV is lower than $1,000 \text{ h}^{-1}$, it will not be economical, because the amount of gas treated is small, and if the GHSV is higher than $200,000 \text{ h}^{-1}$, the time of contact with the catalyst will be short, and thus the adsorption of NO_x or N_2O will decrease.

Meanwhile, the pressure of the gas during passage through the catalyst layer is preferably atmospheric pressure (1 atm) or higher, and an increase in the pressure leads to an increase in the adsorption rate of nitrogen oxides.

According to the present invention, the temperature of the gas during passage through the catalyst layer is 20 to $1,000^\circ\text{C}$, and preferably 200 to 500°C . If the temperature of the gas during passage through the catalyst layer is lower than 20°C , the adsorption, reduction and desorption rates of nitrogen oxides will decrease, and if it is higher than 1000°C , it will thermally damage the catalyst and the support.

Mode for Invention

Hereinafter, the present invention will be described with reference to the following examples in order to more clearly describe the effects of the present invention.

Example 1

A gas comprising 747 ppm of NO_x , 1.69% of O_2 and a balance of nitrogen was passed over 1.285 g of a metal oxide catalyst, prepared from a hydrotalcite precursor and having a molar ratio of $\text{Al}:\text{Pd}:\text{Co} = 1:0.02:1$, at 300°C and a flow rate of 500 ml/min ($\text{SV } 39,000 \text{ hr}^{-1}$) for 55 minutes. As a result, 8.6 mg of NO per g of the catalyst (mg-NO/g-cat.) was adsorbed on the catalyst.

11.1 mg-NO (8.6 mg-NO/g-cat.) adsorbed as described was completely decomposed and desorbed using 4,800 ppm of CO for 30 minutes at 300 °C at a flow rate of 500 ml/min (SV 39,000 hr⁻¹).

5

Example 2

A gas comprising 1,300 ppm of N₂O, 1.48 % of O₂ and a balance of nitrogen was passed over 1.285 g of a metal oxide catalyst, prepared from a hydrotalcite precursor and having a molar ratio of Al:Pd:Co = 1:0.02:1, at
10 300 °C and a flow rate of 500 ml/min (SV 39,000 hr⁻¹) for 50 minutes. As a result, 16.7 mg of N₂O per g of the catalyst (mg-N₂O/g-cat.) was adsorbed on the catalyst.

21.44 mg-N₂O (16.7 mg-N₂O/g-cat.) adsorbed as described was completely decomposed and desorbed using
15 4,850 ppm of CO for 17 minutes at 300 °C at a flow rate of 500 ml/min (SV 39,000 hr⁻¹).

Example 3

A gas comprising 1 % of O₂ and a balance of nitrogen was passed over 2.729 g of a metal oxide
20 catalyst, prepared from a hydrotalcite precursor and having a molar ratio of Al:Pd:Co = 1:0.05:1, at 300 °C and a flow rate of 500 ml/min (SV 20,000 hr⁻¹) for 45 minutes. As a result, 54.4 mg of O₂ per g of the catalyst (mg-O₂/g-cat.) was adsorbed on the catalyst.

25 148.55 mg-O₂ (54.4 mg-O₂/g-cat.) adsorbed as described was completely decomposed and desorbed using 4,000 ppm of CO for 4 hours and 46 minutes at 500 °C at a flow rate of 500 ml/min (SV 20,000 hr⁻¹).

Example 4

30 A gas comprising 484 ppm of NO_x, 1.02 % of O₂ and a balance of nitrogen was passed over 1.294 g of a metal oxide catalyst, prepared from a hydrotalcite precursor and having a molar ratio of Al:Pd:Co = 1:0.05:1, at 300 °C and a flow rate of 500 ml/min (SV 37,500 hr⁻¹) for
35 1 hour. As a result, 7.7 mg of NO per g of the catalyst

(mg-NO/g-cat.) was adsorbed on the catalyst.

9.9 mg-NO (7.7 mg-NO/g-cat.) adsorbed as described was completely decomposed and desorbed using 4,667 ppm of CO at 300 °C for 16 minutes at a flow rate of 500 ml/min (SV 37,500 hr⁻¹).

Example 5

A gas comprising 600 ppm of NO, 1.65 % of O₂ and a balance of nitrogen was passed over 3.684 g of a metal oxide catalyst, prepared from a hydrotalcite precursor and having a molar ratio of Ni:K = 1:0.5, at 300 °C and a flow rate of 500 ml/min (SV 37,500 hr⁻¹) for 5 hours. As a result, 8.5 mg of NO per g of the catalyst (mg-NO/g-cat.) was adsorbed on the catalyst.

31.3 mg-NO (8.5 mg-NO/g-cat.) adsorbed as described was completely decomposed and desorbed using 4,500 ppm of CO at 500 °C for 60 minutes at a flow rate of 500 ml/min (SV 37,500 hr⁻¹).

Example 6

A gas comprising 440 ppm of NO, 1.14 % of O₂ and a balance of nitrogen was passed over 3.72 g of a metal oxide catalyst, prepared from a hydrotalcite precursor and having a molar ratio of Ni:K:Pd = 1:0.2:0.1, at 300 °C and a flow rate of 500 ml/min (SV 33,000 hr⁻¹) for 2 hours. As a result, 1.16 mg of NO per g of the catalyst (mg-NO/g-cat.) was adsorbed on the catalyst.

4.3 mg-NO (1.16 mg-NO/g-cat.) adsorbed as described was desorbed with nitrogen at 500 °C and then completely decomposed using 3,000 ppm of CO for 90 minutes at a flow rate of 500 ml/min (SV 33,000 hr⁻¹).

Example 7

A gas comprising 284 ppm of NO_x, 1.37 % of O₂ and a balance of nitrogen was passed over 3.601 g of a metal oxide catalyst, prepared from a hydrotalcite precursor and having a molar ratio of Ni:Pd:Co = 1:0.1:1, at 300 °C and a flow rate of 700 ml/min (SV 42,000 hr⁻¹) for 45

minutes. As a result, 0.27 mg of NO per g of the catalyst (mg-NO/g-cat.) was adsorbed on the catalyst.

0.98 mg-NO (0.27 mg-NO/g-cat.) adsorbed as described was completely decomposed and desorbed using
5 4,695 ppm of CO at 300 °C for 2 hours and 44 minutes at a flow rate of 700 ml/min (SV 42,000 hr⁻¹).

Example 8

A gas comprising 1,376 ppm of N₂O, 1.48 % of O₂ and a balance of nitrogen was passed over 3.601 g of a metal
10 oxide catalyst, prepared from a hydrotalcite precursor and having a molar ratio of Ni:Pd:Co = 1:0.1:1, at 300 °C and a flow rate of 500 ml/min (SV 30,000 hr⁻¹) for 55 minutes. As a result, 3.25 mg of N₂O per g of the catalyst (mg-N₂O/g-cat.) was adsorbed on the catalyst.

15 11.7 mg-N₂O (3.25 mg-N₂O/g-cat.) adsorbed as described was completely decomposed and desorbed using 4,728 ppm of CO at 300 °C for 1 hour and 5 minutes at a flow rate of 500 ml/min (SV 30,000 hr⁻¹).

As can be seen in the above examples, the
20 catalysts according to the present invention showed high adsorption capacity and had the property of rapidly decomposing nitrogen oxides.

It was reported that Pt-impregnated catalysts based on BaO generally have an adsorption capacity of 10
25 to 20 mg-NO_x/g-cat (at 300~350 °C). In comparison with such catalysts, it can be seen that the catalysts according to the present invention have significantly high adsorption capacity and the property of rapidly decomposing nitrogen oxides using CO.

30 Although the preferred embodiments of the present invention have been described for illustrative purposes, those skilled in the art will appreciate that various modifications, additions and substitutions are possible, without departing from the scope and spirit of the
35 invention as disclosed in the accompanying claims.

Claims

1. A method for decomposing nitrogen oxides using a mixed metal oxide catalyst, the method comprising the steps of:

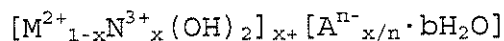
i) providing a catalyst layer comprising a mixed metal oxide catalyst prepared from a hydrotalcite-type or brucite-type compound precursor represented by the following formula 1, wherein the order of adsorption strength of adsorbants thereon is $(\text{NO}, \text{N}_2\text{O}, \text{NO}_2, \text{SO}_2) > \text{CO} > \text{O}_2$, a catalyst wherein a precious metal is impregnated or intercalated into the mixed metal oxide catalyst, or a catalyst wherein a co-catalyst is added and a non-precious metal is impregnated or intercalated into the mixed metal oxide catalyst;

ii) passing a waste gas containing oxygen and nitrogen oxides and having an oxygen concentration of 0.01% or more, through the catalyst layer, to selectively adsorb the nitrogen oxides on the catalyst layer;

iii) passing a reducing agent through the catalyst layer having the nitrogen oxides adsorbed thereon; and

iv) decomposing and desorbing the adsorbed nitrogen oxides by the reducing agent:

[Formula 1]



wherein M^{2+} is selected from the group consisting of Mg^{2+} , Ca^{2+} , Ni^{2+} , Zn^{2+} , Sr^{2+} , Ba^{2+} , Fe^{2+} , Cu^{2+} , Co^{2+} , Pa^{2+} and Mn^{2+} ; N^{3+} is selected from the group consisting of Al^{3+} , Mn^{3+} , Fe^{3+} , Co^{3+} , Ni^{3+} , Cr^{3+} , Ga^{3+} , B^{3+} , La^{3+} , Ce^{3+} and Rh^{3+} ; A^{n-} is selected from the group consisting of CO_3^{2-} , NO_3^- , SO_4^{2-} , Cl^- , OH^- , SiO_3^{2-} , MnO_4^{2-} , HPO_3^{2-} , MnO_4^{2-} , HGao_3^{2-} , HVO_4^{2-} , ClO_4^- and BO_3^{2-} ; x ranges from 0.0 to 0.5; and b is an integer ranging from 0 to 20.

2. The method of claim 1, wherein, SO_2 is desorbed from the catalyst by any one or more of CO , NO , NO_2 , O_2 and N_2 in case that SO_2 is adsorbed on the catalyst such that a decrease in the activity of the catalyst by the accumulation of SO_2 does not occur.

3. The method of claim 1, wherein the precious metal is selected from the group consisting of Mo, Ti, Pt, Au, Ag, Rh, Pa, La, Ir, V, Kr, Nd, Nb, Se, Sc, Ru, In, Y, Z, and mixtures thereof, and the amount of the precious metal in the intercalated catalyst is 0.01-50 wt%.

4. The method of claim 1, wherein the co-catalyst is selected from among alkali metals, and the non-precious metal is selected from the group consisting of Ni, Co, Fe, Mn, Al, Ce, Zr, Cu, Zn, Ba, Mg, Ca, Sr, and mixtures thereof, and the amount of the non-precious metal in the intercalated catalyst is 0.01-99.9 wt%.

5. The method of claim 1, wherein the reducing agent further comprises a component selected from the group consisting of CO , H_2 , $\text{C}_1\text{-C}_6$ hydrocarbons, $\text{C}_1\text{-C}_6$ alcohols, and mixtures thereof.

6. The method of claim 1, wherein the hourly space velocity of the gas during passage through the catalyst layer is $1,000 \text{ h}^{-1}$ to $300,000 \text{ h}^{-1}$.

7. The method of claim 1, wherein the reaction temperature is 20 to 1,000 °C.

8. The method of claim 1, wherein the waste gas comprises 0.001 to 30 wt% of N_2O , 0.001 to 10 wt% of NO, 0.001 to 21 wt% of O_2 , 0.001 to 20 wt% of the reducing agent, and a balance of other contaminants and inert carrier gases.