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(54) Title: CATALYST COMPOSITION AND METHOD FOR USE IN SELECTIVE CATALYTIC REDUCTION OF NITROGEN OXIDES

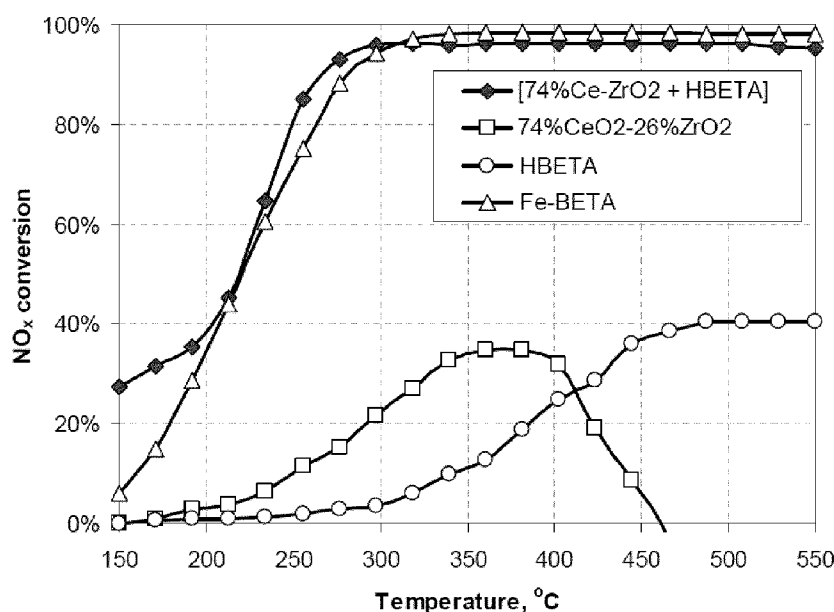


FIG. 1

(57) Abstract: Catalyst composition for selective reduction of nitrogen oxides and soot oxidation comprising a physical mixture of one or more acidic zeolite or zeo-type components with one or more redox active metal compounds and a method for selective reduction of nitrogen oxides and soot oxidation by use of the catalyst composition.



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Title: Catalyst composition and method for use in selective catalytic reduction of nitrogen oxides

5 The present invention relates to catalyst composition for use in selective reduction of nitrogen oxides in off-gases by reaction with ammonia or a precursor thereof.

10 Catalysts for NH_3 -SCR, i.e. selective reduction of nitrogen oxides (NO_x) by use of ammonia as reductant are well known in the art. Those catalysts include zeolitic material, optionally promoted with copper or iron

15 The problem to be solved by this invention is to provide a catalyst composition and method for the reduction of nitrogen oxides with a DeNO_x activity at reaction temperatures between 150 and 550°C.

20 Off-gases from lean combustion engines contain in addition to NO_x , hydrocarbons, CO and soot particles which can be reduced or removed by catalytic oxidation. Consequently, the catalyst composition and method of this invention shall further include soot and hydrocarbon oxidation activity simultaneously with the DeNO_x activity.

25 Our recent studies revealed several examples of a pronounced synergistic effect in composite catalysts prepared by mechanical mixing of acidic zeolite or zeotype powder and redox active metal compounds.

30 We have found that catalyst composition comprising a one or more acidic zeolite or zeotype components physically ad-

mixed with one or more redox active metal compounds shown an improved activity in the selective reduction of nitrogen oxides and oxidation of hydrocarbons, CO and soot contained in off-gas.

5

The term "redox active metal compounds" as used herein relates to metal compounds which reversibly can be oxidized and reduced in terms of changes in oxidation number, or oxidation state, of the metal atom or compound.

10

Pursuant to the above findings, the present invention provides a catalyst composition for selective reduction of nitrogen oxides and soot oxidation comprising one or more acidic zeolite or zeotype components selected from the group consisting of BEA, MFI, FAU, FER, CHA, MOR or mixtures thereof physically admixed with one or more redox active metal compounds selected from the group consisting of Cu/Al₂O₃, Mn/Al₂O₃, CeO₂-ZrO₂, Ce-Mn/Al₂O₃ and mixtures thereof.

20

Catalyst compositions prepared by mechanical mixing of the above mentioned zeolites or zeotype materials and redox metal components mixing according to the invention exhibit a pronounced synergistic effect. DeNO_x activity of such composite catalysts significantly exceeds activity of their individual components.

25

The acidic zeolite or zeotype component can be used in protonic form or promoted with Fe.

30

Preferably, the weight ratio between the zeolite components and the redox components is between 1:1 to 1:50

In an embodiment of the invention, the redox components are dispersed on a support selected from the group consisting of of Al_2O_3 , TiO_2 , SiO_2 , CeO_2 , ZrO_2 or mixtures thereof.

5

It is generally preferred that the mean molar ratio Si/Al of the zeolite components according to the invention is from 5 to 100.

10

The above described catalyst composition according to the invention can be utilised as coating material or as coat on structured bodies of metallic, ceramic, metal oxide, SiC or silica materials or fibres.

15

Thus, the invention provides furthermore a monolithic structured body being coated with a catalyst composition according to anyone of the above disclosed embodiments of the invention.

20

The monolithic structured body is preferably made from metallic, ceramic, metal oxide, SiC or silica fiber materials.

25

The monolithic structured body may be in form of a particle filter, e.g. a honeycomb structured filter or a wall flow filter.

30

In further an embodiment, the catalyst composition is coated on the body in of two or several separate catalyst layers in series or as two or several catalyst layers in parallel and wherein the layers have different compositions or layer thicknesses.

Specific advantages resulting from the invention are

1) Addition of $\text{CeO}_2\text{-ZrO}_2$, $\text{Cu/Al}_2\text{O}_3$, $\text{Mn/Al}_2\text{O}_3$ or $\text{Ce-Mn/Al}_2\text{O}_3$ to acidic zeolite or zeotype in protonic form or promoted with iron markedly enhances DeNO_x activity at $T_{\text{react}} < 250^\circ\text{C}$ without increasing amount of zeolite component. In this case, overall volume of the catalyst is increased by the volume of redox component added.

10

2) Alternatively, amount of expensive zeolite/zeotype component in the composite catalyst can be significantly reduced by its replacement with equivalent volume of redox component. In this case overall volume of the catalyst remains constant, but the amount of zeolite component can be decreased by 2-5 times, without notable sacrificing DeNO_x performance. When $\text{Ce-Mn/Al}_2\text{O}_3$ component is used for the catalyst preparation, notable improvement of NO_x conversion at $T_{\text{react}} < 250^\circ\text{C}$ is observed despite decreased amount of zeolite component.

20

3) In addition to favourable DeNO_x activity, $[\text{CeO}_2\text{-ZrO}_2 + \text{zeolites/zeotypes}]$ or $[\text{Ce-Mn/Al}_2\text{O}_3 + \text{zeolites/zeotypes}]$ compositions demonstrate significant soot oxidation activity, which makes them promising candidates for development of integrated $\text{DeNO}_x\text{-DeSoot}$ catalytic systems.

25

4) In addition to favorable DeNO_x activity, $[\text{CeO}_2\text{-ZrO}_2 + \text{zeolites/zeotypes}]$ or $[\text{Ce-Mn/Al}_2\text{O}_3 + \text{zeolites/zeotypes}]$ compositions demonstrate significantly lower ammonium slip at high temperature due to selective oxidation of excess ammonia.

30

The invention provides additionally a method for the selective reduction of nitrogen oxides and oxidation of soot contained in an off-gas comprising the step of contacting the off-gas in presence of ammonia with a catalyst composition comprising one or more acidic zeolite or zeotype components selected from the group consisting of BEA, MFI, FAU, FER, CHA, MOR or mixtures thereof physically admixed with one or more redox active metal compounds selected from the group consisting of Cu/Al₂O₃, Mn/Al₂O₃, CeO₂-ZrO₂, Ce-Mn/Al₂O₃ and mixtures thereof.

The acidic zeolite or zeotype component can be used in protonic form or promoted with Fe

In an embodiment of the inventive method, the one or more redox active metal compounds are dispersed on a support selected from the group consisting of Al₂O₃, TiO₂, SiO₂, ZrO₂ or mixtures thereof.

In still an embodiment of the inventive method, the catalyst composition is contacted with the off-gas at a temperature below 250°C.

In a further embodiment of the inventive method excess of ammonia is selectively oxidized to nitrogen by contact with the catalyst composition.

Examples

Example 1

5 Synergistic effect in NH_3 -DeNO_x over CeO_2 -ZrO₂ + H-Beta zeolite catalyst compositions.

[CeO_2 -ZrO₂ + H-Beta zeolite] composite catalyst was prepared by thorough mixing 74wt% CeO_2 -26wt%ZrO₂ powder with H-Beta powder at a weight ratio of 10. This weight ratio results in volume ratio of components CeO_2 -ZrO₂/H-Beta = 3/1 due to difference in densities of these materials. The powders were thoroughly grinded in agate mortar for 10-15 min, followed by pelletization. The pellets were crushed and sieved collecting 0.2 - 0.4 mm fraction for catalytic test. Similarly pelletized 74wt% CeO_2 -26wt%ZrO₂, H-Beta, and Fe-Beta zeolite were used as reference samples.

The catalysts were tested in the NH_3 -DeNO_x in the temperature range of 150-550 °C. The test was performed under following conditions: decreasing reaction temperature with a rate of 2°C/min, feed gas composition: 500 ppm NO, 540 ppm NH_3 , 10 vol % O₂, 6 vol% H₂O, balanced with N₂ to obtain a total flow of 300 mL/min.

Catalyst loading and resulted GHSV:

0.197g with 74wt% CeO_2 -ZrO₂ + 0.02g H-Beta zeolite, catalyst volume 0.134 ml, GHSV = 135 000 h⁻¹

Under these conditions CeO_2 -ZrO₂ + H-Beta zeolite composite catalyst showed DeNO_x activity, which substantially

exceeded activities of individual 74wt%CeO₂-ZrO₂ (0.131g CeO₂-ZrO₂, catalyst volume 0.067 ml, GHSV = 270,000 h⁻¹) and H-Beta zeolite (0.04g, catalyst volume 0.067 ml, GHSV = 270 000 h⁻¹), indicating pronounced synergistic effect between components of composite catalyst as shown in Fig. 1.

NO_x conversion over composite catalyst is similar to NO_x conversion over commercial Fe-Beta zeolite (Fe-Beta) at 230-550°C, and exceeds NO_x conversion over Fe-Beta zeolite at 150-200°C.

Example 2

Enhanced DeNO_x performance of [CeO₂-ZrO₂ + Fe-Beta] composite catalyst at T_{react} < 250°C

Two samples of [CeO₂-ZrO₂ + Fe-Beta zeolite] composite catalyst were prepared by thorough grinding of 74wt%CeO₂-26wt%ZrO₂ and Fe-Beta zeolite powders.

A first sample was prepared by mixing 74wt%CeO₂-26wt%ZrO₂ and Fe-Beta zeolite powders at a weight ratio of 3.3. This weight ratio results in a volume ratio of 74wt%CeO₂-26wt%ZrO₂/Fe-Beta components in composite catalyst = 1/1.

A second sample was prepared by mixing 74wt%CeO₂-26wt%ZrO₂ and Fe-Beta powders at a weight ratio of 10. For the second sample volume ratio of 74wt%CeO₂-26wt%ZrO₂/Fe-Beta zeolite equals 3/1.

After grinding in agate mortar for 10-15 min, the resulted mixtures were pelletized. The pellets were crushed and

sieved collecting 0.2 - 0.4 mm fraction for catalytic test. Similarly pelletized Fe-Beta zeolite was used as reference.

Activities of the prepared samples were tested using the following catalyst loading which kept constant amount of Fe-Beta zeolite component in the reactor:

The first sample with 1/1 volume component ratio: [0.065g 74%CeO₂-ZrO₂ + 0.02g Fe-Beta zeolite].

The second sample with 3/1 volume component ratio: [0.197g 74%CeO₂-ZrO₂ + 0.02g Fe-Beta zeolite].

Reference sample: 0.02 g Fe-Beta zeolite.

The catalysts were tested in NH₃-DeNO_x within the temperature range of 150-550 °C. The test was performed under following conditions: decreasing reaction temperature with a rate of 2°C/min, feed gas composition: 500 ppm NO, 540 ppm NH₃, 10 vol % O₂, 6 vol% H₂O, balanced with N₂ to obtain a total flow of 300 mL/min.

Catalyst loading and resulted GHSV:

[0.197g 74%CeO₂-ZrO₂ + 0.02g Fe-Beta zeolite], catalyst vol. = 0.134 ml, GHSV = 135 000 h⁻¹;

[0.065g 74%CeO₂-ZrO₂ + 0.02g Fe-Beta zeolite], catalyst vol. = 0.067 ml, GHSV = 270 000 h⁻¹;

0.02 Fe-Beta zeolite, catalyst vol. = 0.034 ml, GHSV = 540 000 h⁻¹.

Under these test conditions [CeO₂-ZrO₂ + Fe-Beta zeolite] composite catalysts showed enhanced DeNO_x activity within

low-temperature range (150-300°C), which significantly exceeded activity of individual Fe-Beta zeolite, as shown in Fig. 2. It is important to note that the activity of [CeO₂-ZrO₂ + Fe-Beta zeolite] is improved when the amount of
5 CeO₂-ZrO₂ component was increased.

Example3

Catalyst with reduced amount of zeolite component.

10

Three samples of [CeO₂-ZrO₂ + Fe-Beta zeolite] composite catalyst were prepared by thorough grinding of 74wt%CeO₂-26wt%ZrO₂ powder with Fe-Beta zeolite powder:

15 A first sample was prepared by mixing 74wt%CeO₂-26wt%ZrO₂ and Fe-Beta powders at a weight ratio of 3.3. In this case volume ratio of 74wt% CeO₂- 26wt% ZrO₂ /Fe-Beta zeolite equals 1/1.

20 A second sample was prepared by mixing 74wt%CeO₂-26wt%ZrO₂ and Fe-Beta zeolite powders at a weight ratio of 15.5. For the second sample volume ratio of 74wt%CeO₂-26wt%ZrO₂ and Fe-Beta zeolite components equals 5/1.

25 A third sample was prepared by was prepared by mixing 74wt%CeO₂-26wt%ZrO₂ and Fe-Beta zeolite powders at a weight ratio of 30. For the second sample volume ratio of 74wt%CeO₂-26wt%ZrO₂ and Fe-Beta zeolite components equals 10/1.

30

After grinding in agate mortar for 10-15 min, the resulted mixtures were pelletized. The pellets were crushed and

sieved collecting 0.2 - 0.4 mm fraction for catalytic test. Similarly pelletized Fe-Beta zeolite was used as reference.

Activities of the prepared samples were tested using the following catalyst loading which kept constant volume of the catalyst in the reactor. In all experiments described below overall volume on the catalyst loaded was 0.067 ml, which results in GHSW $\sim 270\ 000\ \text{h}^{-1}$:

First sample (1/1 vol component ratio): [0.065g 74wt%CeO₂-ZrO₂ + 0.02g Fe-Beta zeolite].

Second sample (5/1 vol component ratio): [0.109g 74wt%CeO₂-ZrO₂ + 0.007g Fe-Beta zeolite].

Third sample (10/1 vol component ratio): [0.119g 74wt%CeO₂-ZrO₂ + 0.0035g Fe-Beta zeolite].

Reference sample: 0.02 g Fe beta-zeolite.

Feed gas composition: 540 ppm NH₃, 500 ppm NO, 10 % O₂, 6 % H₂O balance with N₂.

Under these conditions [CeO₂-ZrO₂ + Fe-Beta zeolite] composite catalysts showed DeNO_x performances, which were essentially identical to the performance of reference Fe-Beta zeolite sample, despite significantly reduced amount of zeolite catalyst (Fe-Beta zeolite) loaded into the reactor as a part of composite [CeO₂-ZrO₂ + Fe-Beta zeolite].

The data in Fig. 3 show that amount of zeolite can be reduced at least 10 times without sacrificing DeNO_x perform-

ance of [CeO₂-ZrO₂ + Fe-Beta zeolite] by its replacement with corresponding volume of CeO₂-ZrO₂.

Example 4

5

Enhanced DeNO_x performance of [Ce-Mn/Al₂O₃ + Fe-Beta zeolite] composite catalyst at T_{react} < 250 °C.

[Ce-Mn/Al₂O₃ + Fe-Beta] composite catalysts were prepared by thorough mixing 15wt%Ce-15wt%Mn/Al₂O₃ powder with Fe-Beta powder at a weight ratio of 0,8:1; 1,7:1 and 3,4:1 keeping the same total volume of the catalyst constant. These weight ratios result in volume ratio of components Ce-Mn/Al₂O₃/Fe-Beta = 2/1; 1/1 and 1/2 due to difference in densities of these materials. The powders were thoroughly grinded in agate mortar for 10-15 min, followed by pelletization. The pellets were crushed and sieved collecting 0.2 - 0.4 mm fraction for catalytic test. Similarly pelletized Fe-Beta was used as reference.

20

The catalysts were tested in the NH₃-DeNO_x in the temperature range of 150-550 °C. The test was performed under following conditions: decreasing reaction temperature with a rate of 2°C/min, feed gas composition: 500 ppm NO, 540 ppm NH₃, 10 vol % O₂, 6 vol% H₂O, balanced with N₂ to obtain a total flow of 300 mL/min.

25

Catalyst load: 0.04g Fe-Beta and [0.045g Ce-Mn/Al₂O₃ + 0.013g Fe-Beta] (2/1 ratio), [0.034g Ce-Mn/Al₂O₃ + 0.02g Fe-Beta] (1/1 ratio), [0.022g Ce-Mn/Al₂O₃ + 0.027g Fe-Beta] (1/2 ratio).

30

Under these conditions all [Ce-Mn/Al₂O₃ + Fe-Beta] composite catalysts showed DeNO_x activity, which radically exceeded activities of individual Ce-Mn/Al₂O₃ and Fe-Beta at temperatures below 350°C, indicating pronounced synergistic effect between components of composite catalyst (Fig. 4). Besides that, ammonia slip on composite catalysts was significantly lower than for a reference Fe-Beta catalyst indicating that those composite systems can be used as integrated DeNO_x-ASC.

Example 5

Enhanced DeNO_x performance of [10wt%Cu/Al₂O₃ + H-zeolite] composite catalysts.

Three samples of [10wt%Cu/Al₂O₃ + H-zeolite] composite catalyst were prepared by thorough grinding of 10wt%Cu/Al₂O₃ and H-Beta, H-ZSM-5, or H-ferrierite powder.

A first sample was prepared by mixing 10wt%Cu/Al₂O₃ and H-Beta (Si/Al = 20) powders at a weight ratio of 1/1.

A second sample was prepared by mixing 10wt%Cu/Al₂O₃ and H-ZSM-5 powders (Si/Al = 20) at a weight ratio of 1/1.

A third sample was prepared by mixing 10wt%Cu/Al₂O₃ and H-ferrierite powders (Si/Al = 32) at a weight ratio of 1/1.

After grinding in agate mortar for 10-15 min, the resulted mixtures were pelletized. The pellets were crushed and sieved collecting 0.2 - 0.4 mm fraction for catalytic test.

Similarly corresponding pelletized zeolites (H-Beta, H-ZSM-5, and H-ferrierite) were used as reference.

Activities of the prepared samples were tested using the following catalyst loading which kept constant amount of zeolite component in the reactor:

The first sample with 1/1 weight component ratio: [0.040g 10wt%Cu/Al₂O₃ + 0.040g H-Beta].

The second sample with 1/1 weight component ratio: [0.040g 10wt%Cu/Al₂O₃ + 0.040g H-ZSM-5].

The third sample with 1/1 weight component ratio: [0.040g 10wt%Cu/Al₂O₃ + 0.040g H-ferrierite].

Reference samples: 0.040 g H-Beta; 0.040g H-ZSM-5, or H-ferrierite, or 0.040 g 10wt%Cu/Al₂O₃.

The catalysts were tested in NH₃-DeNO_x within the temperature range of 150-550 °C. The test was performed under following conditions: decreasing reaction temperature with a rate of 2°C/min, feed gas composition: 500 ppm NO, 540 ppm NH₃, 10 vol % O₂, 6 vol% H₂O, balanced with N₂ to obtain a total flow of 300 mL/min.

Catalyst loading and resulted GHSV:

[0.040g 10wt%Cu/Al₂O₃ + 0.040g H-Beta], catalyst vol. = 0.134 ml, GHSV = 135 000 h⁻¹;

[0.040g 10wt%Cu/Al₂O₃ + 0.040g H-ZSM-5], catalyst vol. = 0.134 ml, GHSV = 135 000 h⁻¹;

[0.040g 10wt%Cu/Al₂O₃ + 0.040g H-ferrierite], catalyst vol. = 0.134 ml, GHSV = 135 000 h⁻¹;

Reference catalysts

0.040g H-Beta, catalyst vol. = 0.067 ml,

5 GHSV = 270,000 h⁻¹;

0.040g H-ZSM-5, catalyst vol. = 0.067 ml,

GHSV = 270,000 h⁻¹;

0.040g H-ferrierite, catalyst vol. = 0.067 ml,

GHSV = 270,000 h⁻¹;

10 0.040g Cu/Al₂O₃, catalyst vol. = 0.067 ml,

GHSV = 270,000 h⁻¹.

Under these test conditions [10wt%Cu/Al₂O₃ + H-zeolite] composite catalysts showed enhanced DeNO_x within the whole temperature range (150-550°C), which significantly exceeded activity of individual components, as shown by comparing Fig. 5 and Fig. 6.

Example 6

20

Catalyst with enhanced soot oxidation activity.

[CeO₂-ZrO₂ + Fe-Beta] with 3/1 vol. component ratio was prepared as described in Example 2. For testing soot oxidation activity of [CeO₂-ZrO₂ + Fe-Beta] a part of pelletized sample was crushed, and the catalyst powder was mixed with soot ("Printex U", Degussa) at a weight ratio catalyst/soot = 1/10. Soot and catalyst were mixed by shaking in a glass bottle for 5 min, thus establishing loose contact between soot and the catalyst. Reference sample was prepared in a similar manner using Fe-Beta powder.

30

Soot oxidation was carried out at temperature ramp =
10°C/min in a flow of dried air. Profiles of soot oxidation
over [CeO₂-ZrO₂ + Fe-Beta] and Fe-Beta are displayed in
Fig. 7. [CeO₂-ZrO₂ + Fe-Beta] significantly higher activity
5 in soot oxidation than individual Fe-Beta, as evidenced by
a shift of soot oxidation maximum from ~ 600°C for (Fe-Beta
+ soot) to ~ 420°C for ([CeO₂-ZrO₂ + Fe-Beta] + soot).

Claims:

1. Catalyst composition for selective reduction of nitrogen oxides and soot oxidation comprising of one or more acidic zeolite or zeotype components selected from the group consisting of BEA, MFI, FAU, FER, CHA, MOR or mixtures thereof physically admixed with one ore more redox active metal compounds selected from the group consisting of Cu/Al₂O₃, Mn/Al₂O₃, CeO₂-ZrO₂, Ce-Mn/Al₂O₃ and mixtures thereof.
2. The catalyst composition of claim 1, wherein weight ratio between the zeolite components and the redox components is between 1:1 and 1:50.
3. The catalyst composition of claim 1 or 2, wherein the one or more redox active metal compounds are dispersed on a support selected from the group consisting of of Al₂O₃, TiO₂, SiO₂, ZrO₂ or mixtures thereof.
4. The catalyst composition according to anyone of claims 1 to 3, wherein the one or more acidic zeolite or zeotype components are in protonic form or promoted with Fe.
5. The catalyst composition according to anyone of claims 1 to 4, wherein mean molar ratio of Si/Al of the one or more acidic zeolite or zeotype components is from 5 to 100.
6. The catalyst composition according to anyone of the preceding claims, wherein the one or more acidic zeolite or zeotype components are selected from the group consisting of beta-zeolite, ZSM-5 and ferrierite.

7. A monolithic structured body being coated with a catalyst composition according to anyone of the preceding claims.

5 8. The monolithic structured body of claim 7, wherein the monolithic structured body is in a form of a particle filter.

10 9. The monolithic structured body of claims 7 or 8, wherein the catalyst composition is coated on the body in two or several separate catalyst layers in series or as two or several catalyst layers in parallel and wherein the layers have different compositions or layer thicknesses.

15 10. Method for the selective reduction of nitrogen oxides and oxidation of soot contained in an off-gas comprising the step of contacting the off-gas in presence of ammonia with a catalyst composition comprising one or more acidic zeolite or zeotype components selected from the group consisting of BEA, MFI, FAU, FER, CHA, MOR or mixtures thereof
20 physically admixed with one or more redox active metal compounds selected from the group consisting of Cu/Al₂O₃, Mn/Al₂O₃, CeO₂-ZrO₂, Ce-Mn/Al₂O₃ and mixtures thereof.

25 11. The method according claim 10, wherein the one or more redox active metal components dispersed on the surface of the one or more zeolite components contain Ce, Mn, Zr, Cr or mixtures thereof.

30 12. The method of according to claim 10 or 11, wherein the catalyst composition is contacted with the off-gas at a temperature below 250°C.

13. The method according to anyone of claim 10 to 12,
wherein excess of ammonia is selectively oxidized to nitro-
gen by contact with the catalyst composition.

5

14. The method according to anyone of claims 10 to 13,
wherein the one or more acidic zeolite or zeotype compo-
nents are in protonic form or promoted with Fe.

10

15. The method according to anyone of claims 10 to 14,
wherein mean molar ratio of Si/Al of the one or more acidic
zeolite or zeotype components is from 5 to 100.

15

16. The method according to anyone of claims 10 to 15,
wherein the one or more acidic zeolite or zeotype compo-
nents are selected from the group consisting of beta-
zeolite, ZSM-5 and ferrierite.

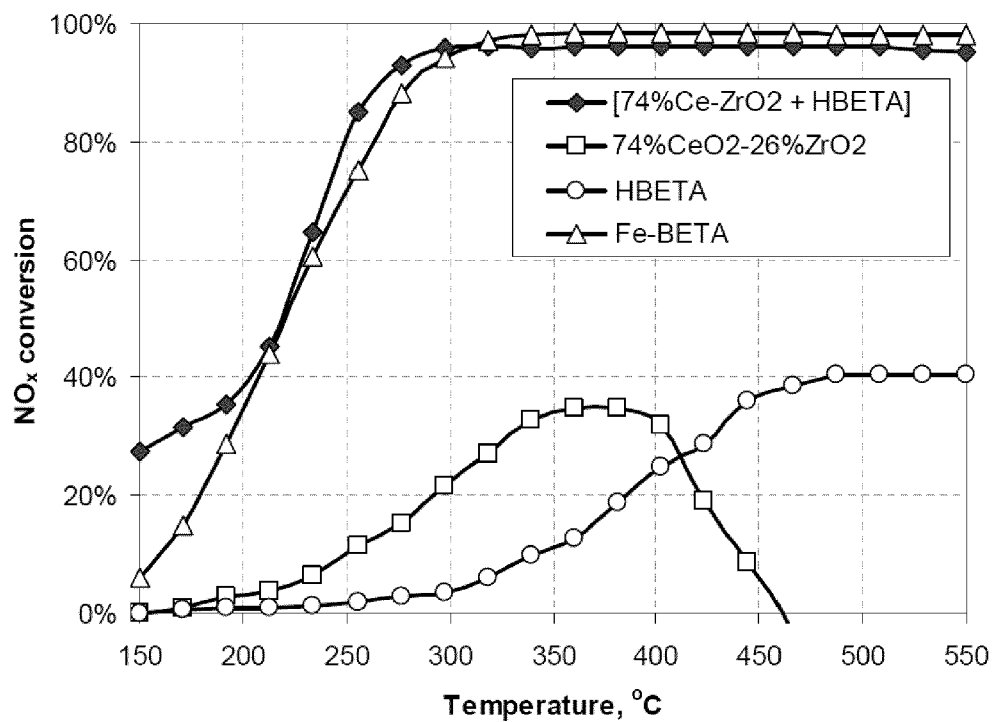


FIG. 1

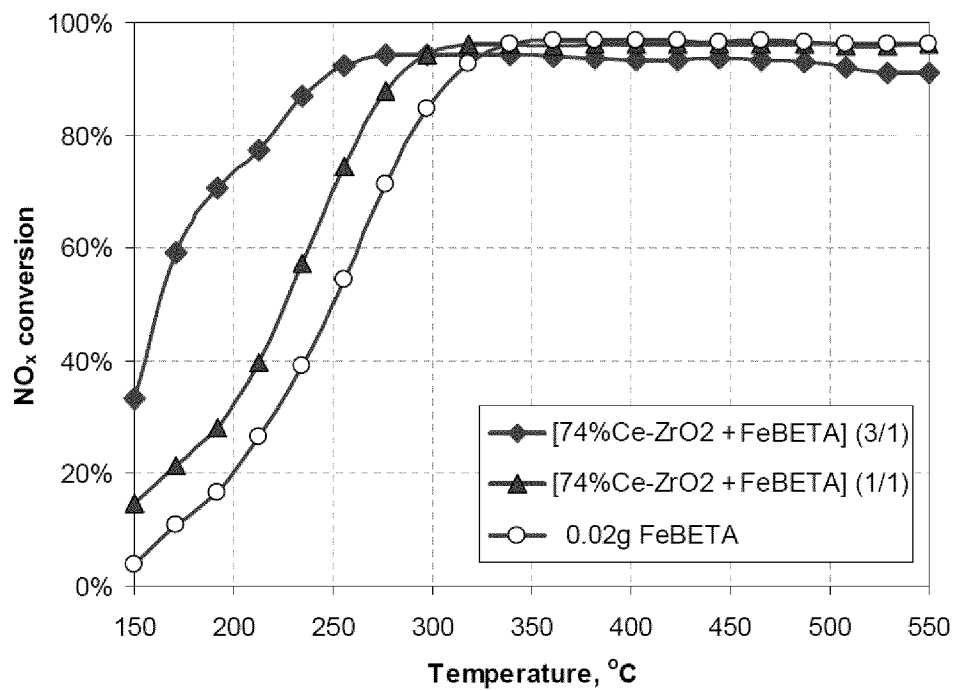


FIG. 2

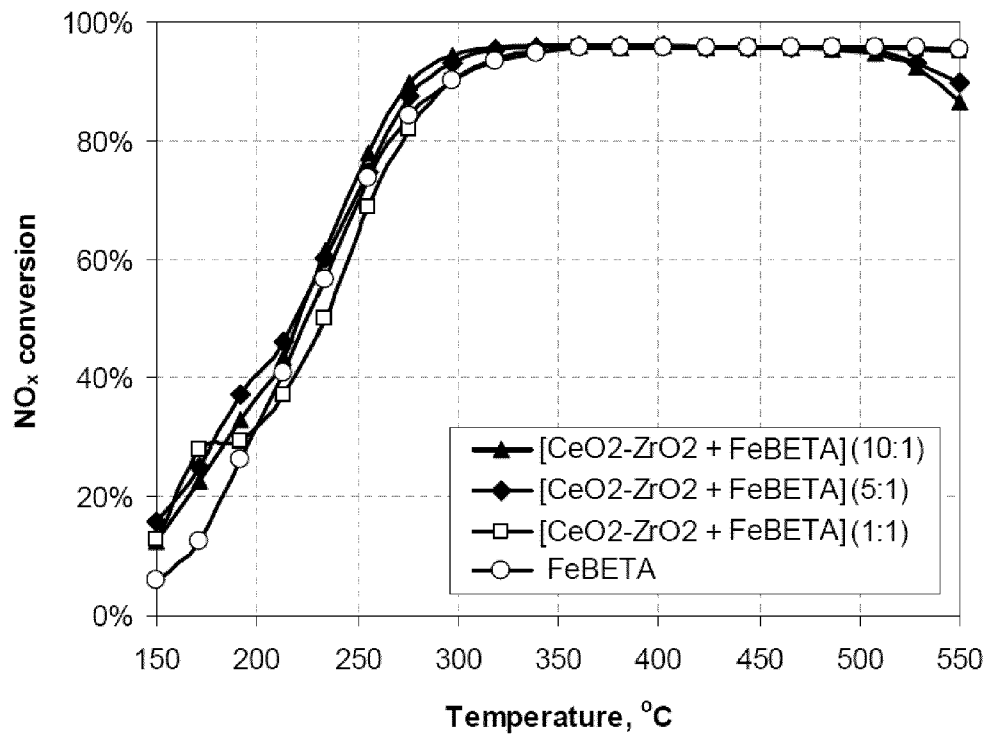


FIG. 3

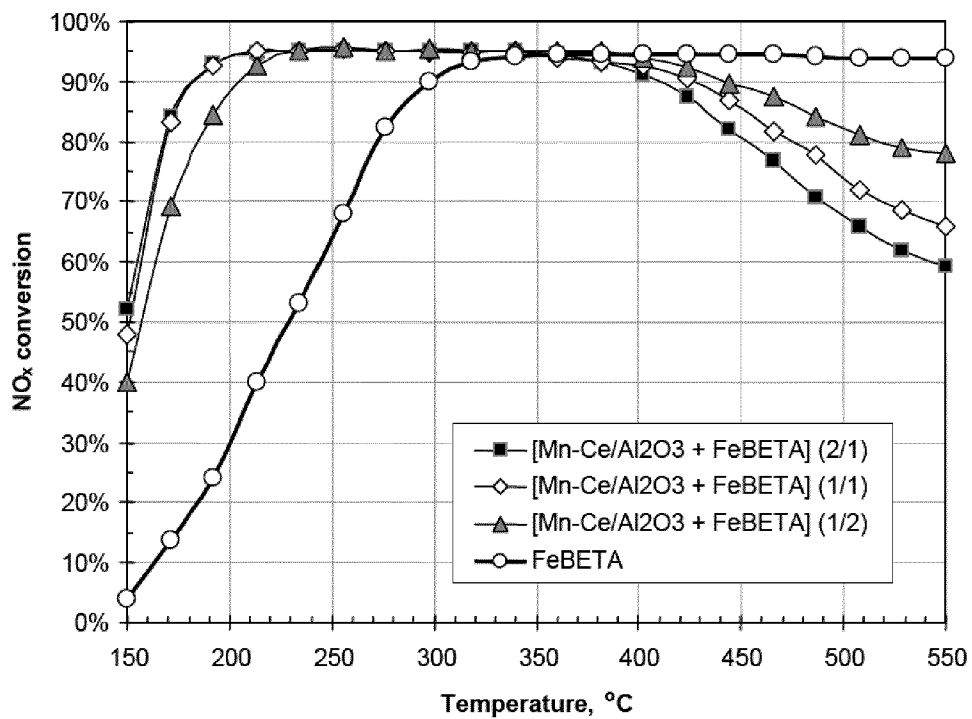


FIG. 4

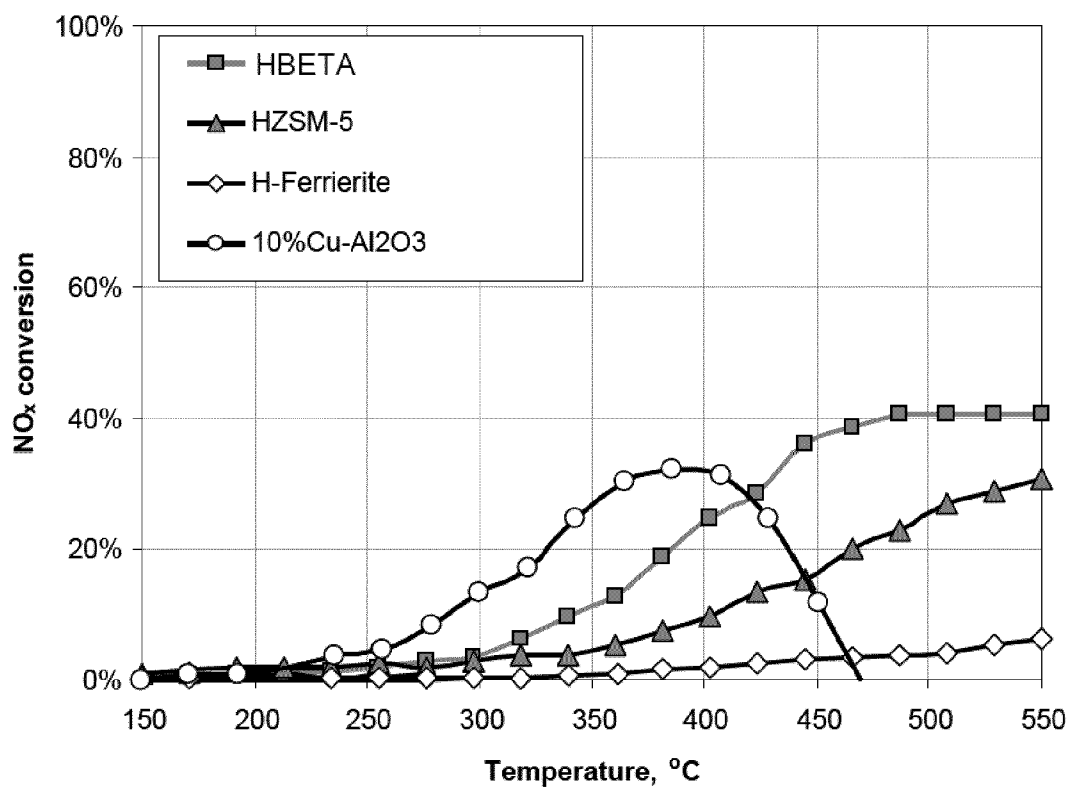


FIG. 5

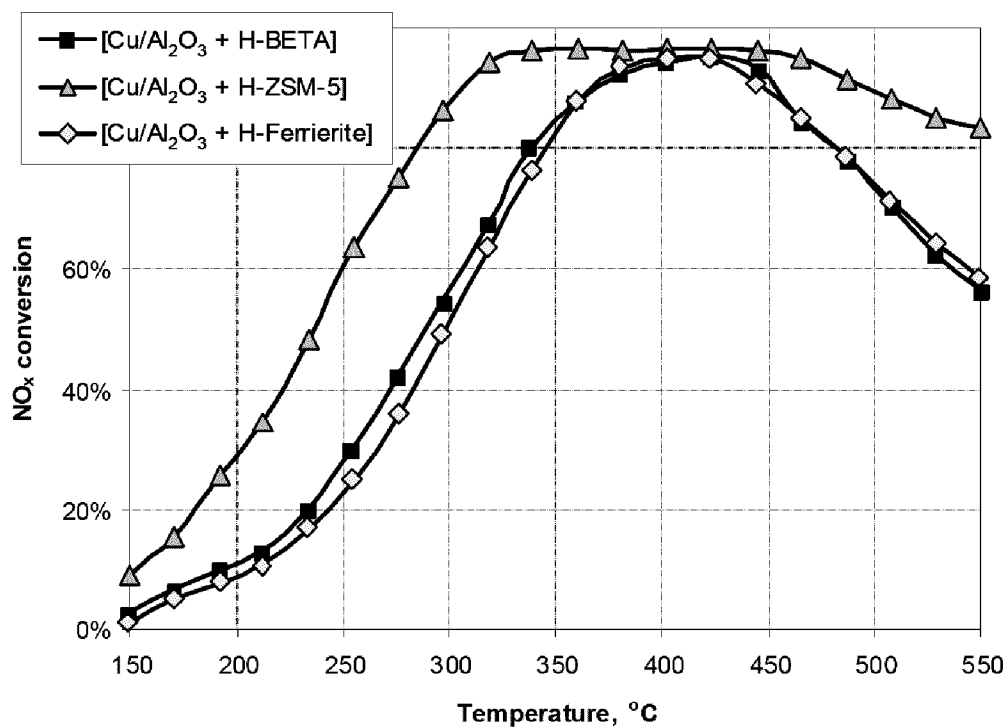
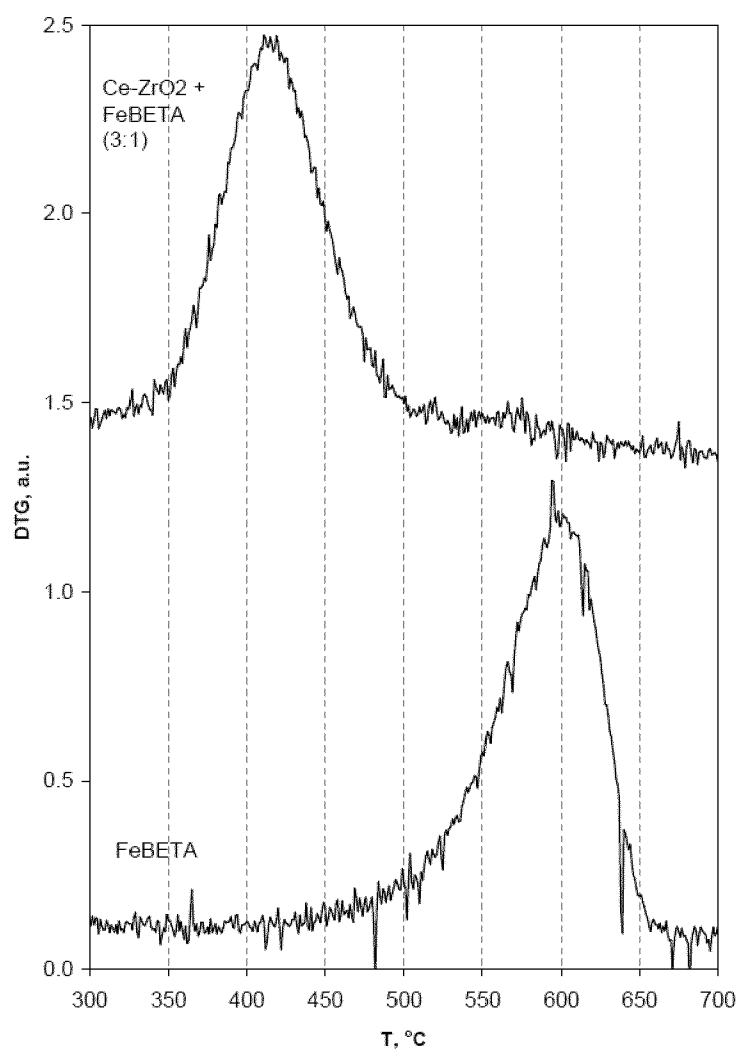


FIG. 6

**FIG. 7**