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(54) Titre : MATERIAUX ZEOLITIQUES ACTIVES PAR LE CUIVRE DE LA STRUCTURE DE CADRE CHA ISSUS D'UNE SYNTHESE SANS MODELE ORGANIQUE, ET LEUR UTILISATION EN REDUCTION CATALYTIQUE SELECTIVE DE NOX

(54) Title: COPPER-PROMOTED ZEOLITIC MATERIALS OF THE CHA FRAMEWORK STRUCTURE FROM ORGANOTEMPLATE-FREE SYNTHESIS AND USE THEREOF IN THE SELECTIVE CATALYTIC REDUCTION OF NOX

(57) **Abrégé/Abstract**:

A synthetic copper and/or iron containing zeolitic material having a CHA framework structure, wherein the CHA framework structure comprises $SiO_2$,$X_2O_3$, and optionally comprises $Z_2O_5$, wherein X is a trivalent element, and Z is a pentavalent element, wherein the zeolitic material contains from 3.8 to 12 wt.% of Cu and/or Fe calculated as the respective element and based on 100 wt.% of $SiO_2$ contained in the zeolitic material having a CHA framework structure, and wherein the $^{29}$Si MAS NMR of the zeolitic material comprises: a first peak (P1) in the range of from -96 to -98.8 ppm; a second peak (P2) in the range of from -102 to -104.5 ppm; and a third peak (P3) in the range of from -107.5 to -111 ppm; wherein the integration of the first, second, and third peaks in the $^{29}$Si MAS NMR of the zeolitic material offers a ratio of the integration values P1:P2:P3 ranging from (0.35-0.7):1: (0.1-1.6), as well as to a process for its preparation, and to the use thereof, in particular in a method for the selective catalytic reduction of $NO_x$.

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**(54) Title:** COPPER-PROMOTED ZEOLITIC MATERIALS OF THE CHA FRAMEWORK STRUCTURE FROM ORGANOTEMPLATE-FREE SYNTHESIS AND USE THEREOF IN THE SELECTIVE CATALYTIC REDUCTION OF $NO_X$

**(57) Abstract:** A synthetic copper and/or iron containing zeolitic material having a CHA framework structure, wherein the CHA framework structure comprises $SiO_2$,$X_2O_3$, and optionally comprises $Z_2O_5$, wherein X is a trivalent element, and Z is a pentavalent element, wherein the zeolitic material contains from 3.8 to 12 wt.% of Cu and/or Fe calculated as the respective element and based on 100 wt.% of $SiO_2$ contained in the zeolitic material having a CHA framework structure, and wherein the $^{29}$Si MAS NMR of the zeolitic material comprises: a first peak (P1) in the range of from -96 to -98.8 ppm; a second peak (P2) in the range of from -102 to -104.5 ppm; and a third peak (P3) in the range of from -107.5 to -111 ppm; wherein the integration of the first, second, and third peaks in the $^{29}$Si MAS NMR of the zeolitic material offers a ratio of the integration values P1:P2:P3 ranging from (0.35-0.7):1: (0.1-1.6), as well as to a process for its preparation, and to the use thereof, in particular in a method for the selective catalytic reduction of $NO_x$.

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# COPPER-PROMOTED ZEOLITIC MATERIALS OF THE CHA FRAMEWORK STRUCTURE FROM ORGANOTEMPLATE-FREE SYNTHESIS AND USE THEREOF IN THE SELECTIVE CATALYTIC REDUCTION OF $NO_X$

## TECHNICAL FIELD

The present invention relates to an organotemplate-free synthetic process for the production of a zeolitic material having a CHA framework structure ion-exchanged with high levels of copper and/or iron and to the copper and/or iron exchanged zeolitic material as obtained or obtainable according to the inventive method as well as to the synthetic copper and/or iron containing zeolitic material per se and to its use in a method for the treatment of $NO_x$ by selective catalytic reduction (SCR). Furthermore, the present invention relates to the use of the synthetic copper and/or iron containing zeolitic material, in particular as a catalyst in the treatment of $NO_x$ containing automotive or industrial exhaust gas by SCR.

## INTRODUCTION

Zeolites containing copper and/or iron have found wide use in the field of selective catalytic reduction of nitrogen oxides ($NO_x$) contained in exhaust gases, and in particular in exhaust gas stemming from diesel engines and lean burn gasoline engines. A particularly prominent example of the zeolites which find use in these applications are copper containing zeolites of the CHA structure type, and in particular chabazite ion-exchanged with copper.

Thus, WO 2009/141324 A1, for example, relates to a process for the direct synthesis of Cu containing zeolites having CHA structure and to their use in the selective catalytic reduction of $NO_x$ in exhaust gas.

WO 2013/068976 A1, on the other hand, concerns an organotemplate-free synthetic process for the production of a zeolitic material of the CHA-type framework structure.

In view of increasingly restrictive emissions regulations for $NO_x$ emissions, in particular in the field of automotive exhaust gas emissions, there is however an ongoing need for the provision of more efficient calayst materials which are active in the fresh and aged states in order to meet current and future requirements and regulations. In particular, there is a need for new zeolitic materials which in combination with catalytic metals provide levels of exhaust gas treatment efficiencies which compare to the levels achieved by the current bench-mark catalysts yet may be obtained at lower cost and/or which outperform the currently used materials such as copper chabazite, on particular with respect to their aptitude to reduce $NO_x$ via selective catalytic reduction.

## DETAILED DESCRIPTION

It is therefore an object of the present invention to provide an improved zeolitic material and in particular an improved zeolite catalyst. More specifically, the present invention aims at providing a zeolite catalyst displaying higher activities under prolonged conditions of use, in particular with respect to applications in selective catalytic reduction (SCR), i.e. for the conversion of nitrogen oxide with a reducing agent to environmentally inoffensive compounds such as in particular to nitrogen and oxygen.

Thus, it has surprisingly been found that zeolitic materials supporting copper and/or iron, wherein said zeolitic materials have a CHA-type framework structure as may be obtained from organotemplate-free synthesis display an improved performance as a catalyst compared to conventional zeolitic materials having the CHA-type framework structure obtained from organotemplate-mediated synthesis, wherein this improved performance is particularly apparent in the selective catalytic reduction reaction. Even more surprisingly, it has been observed that zeolitic materials having a CHA-type framework structure obtainable from organotemplate-free synthetic procedures may not only display a higher catalytic activity at comparable loadings relative to the known catalysts from the prior art wherein the zeolitic material is obtained from templated synthesis, it has furthermore quite unexpectedly been found that said improved catalytic activity observed for the zeolitic materials obtainable from organotemplate-free synthesis may actually be improved towards higher loadings compared to the general decrease of catalytic activity towards higher loadings observed for the prior art materials. All the more surprisingly, said considerably improved catalytic performance is not only observed at particularly low temperatures, but is furthermore sustained even after severe conditions of use as may for example be simulated by hydrothermal aging of the materials, in particular when compared to the known catalytic materials of the prior art.

Therefore, the present invention relates to an organotemplate-free synthetic process for the production of a copper and/or iron containing zeolitic material having a CHA framework structure comprising $YO_2$, $X_2O_3$, and optionally comprising $Z_2O_5$, wherein said process comprises:

(1) preparing a mixture comprising one or more sources for $YO_2$, one or more sources for $X_2O_3$, and seed crystals having a CHA framework structure, wherein the CHA framework structure of the seed crystals comprises $YO_2$, $X_2O_3$, and optionally comprises $Z_2O_5$;

(2) crystallizing the mixture prepared in (1) for obtaining a zeolitic material having a CHA framework structure; and

(3) subjecting the zeolitic material obtained in (2) to one or more ion exchange procedures for obtaining a zeolitic material ion exchanged with Cu and/or Fe, preferably with Cu;

wherein Y is a tetravalent element, X is a trivalent element, and Z is a pentavalent element, wherein the ion exchanged zeolitic material obtained in (3) contains from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $YO_2$ contained in the zeolitic material having a CHA framework structure.

Thus, according to the inventive process, at no point does the mixture prepared in (1) and crystallized in (2) contain more than an impurity of an organic structure directing agent specifically used in the synthesis of zeolitic materials having a CHA-type framework structure, in particular specific tetraalkylammonium compounds, dialkyl amines, heterocyclic amines, and combinations of two or more thereof. Thus, by way of example, at no point does the mixture prepared in (1) and crystallized in (2) contain more than an impurity of one or more organic structure directing agents selected from the group consisting of tetra($C_1$ – $C_5$)alkylammonium compounds, di($C_1$ – $C_5$)alkyl amines, oxygen containing heteroxyclic amines with 5 to 8 ring members, and combinations of two or more thereof, more preferably from the group consisting of tetra($C_2$ – $C_4$)alkylammonium compounds, di($C_2$ – $C_4$)alkyl amines, oxygen containing heteroxyclic amines with 5 to 7 ring members, and combinations of two or more thereof, more preferably from the group consisting of tetra($C_2$ – $C_3$)alkylammonium compounds, di($C_2$ – $C_3$)alkyl amines, oxygen containing heteroxyclic amines with 5 or 6 ring members, and combinations of two or more thereof, and/or related organotemplates such as any suitable N-alkyl-3-quinuclidinol compound, N,N,N-trialkyl-exoaminonorbornane compound, N,N,N-trimethyl-1-adamantylammonium compound, N,N,N-trimethyl-2-adamantylammonium compound, N,N,N-trimethylcyclohexylammonium compound, N,N-dimethyl-3,3-dimethylpiperidinium compound, N,N-methylethyl-3,3-dimethylpiperidinium compound, N,N-dimethyl-2-methylpiperidinium compound, 1,3,3,6,6-pentamethyl-6-azonio-bicyclo(3.2.1)octane compound, N,N-dimethylcyclohexylamine compound, or any suitable N,N,N-trimethylbenzylammonium compound, including combinations of two or more thereof. As noted above, such an impurity can, for example, be caused by organic structure directing agents still present in seed crystals used in the inventive process. Organic structure directing agents eventually contained in seed crystals may not, however, participate in the crystallization process since they are trapped within the seed crystal framework and therefore may not act as structure directing agents within the meaning of the present invention.

Within the meaning of the present invention, an “organotemplate-free” synthetic process relates to a synthetic process wherein the materials used therein are substantially free of organic structure directing agents, wherein “substantially” as employed in the present invention with respect to the amount of one or more organic structure directing agents contained in the one or more materials used in a synthetic process indicates an amount of 0.1 wt.-% or less of one or more organic structure directing agents based on 100 wt.-% of the mixture prepared in (1), preferably 0.05 wt.-% or less, more preferably 0.001 wt.-% or less, more preferably 0.0005 wt.-% or less, and even more preferably 0.0001 wt.-% or less thereof. Said amounts of one or more organic structure directing agents, if at all present in any one of the materials used in the synthetic process, may also be denoted as “impurities” or “trace amounts” within the meaning of the present invention. Furthermore, it is noted that the terms “organotemplate” and “organic structure directing agent” are synonymously used in the present application.

Thus, the term “organotemplate” as employed in the present application designates any conceivable organic material which is suitable for template-mediated synthesis of a zeolite material, preferably of a zeolite material having a CHA-type framework-structure, and even more preferably which is suitable for the synthesis of Chabazite. Such organotemplates include e.g. any suitable tetraalkylammonium compound, dialkyl amine, heterocyclic amine, N-alkyl-3-quinuclidinol compound, N,N,N-trialkyl-exoaminonorbornane compound, N,N,N-trimethyl-1-adamantylammonium compound, N,N,N-trimethyl-2-adamantylammonium compound, N,N,N-trimethylcyclohexylammonium compound, N,N-dimethyl-3,3-dimethylpiperidinium compound, N,N-methylethyl-3,3-dimethylpiperidinium compound, N,N-dimethyl-2-methylpiperidinium compound, 1,3,3,6,6-pentamethyl-6-azonio-bicyclo(3.2.1)octane compound, N,N-dimethylcyclohexylamine compound, and any suitable N,N,N-trimethylbenzylammonium compound.

Therefore, according to the present invention, the preparation of a zeolitic material according to the process defined in (1) and (2) and preferably according to the inventive process as defined in particular and preferred embodiments of the present application is conducted in the absence of an organic structure directing agent and is thus an organotemplate-free synthetic process within the meaning of the present invention.

Furthermore, within the meaning of the present invention, $YO_2$, $X_2O_3$, and optionally $Z_2O_5$ are comprised in the CHA framework structure as structure building elements, as opposed to non-framework elements which can be present in the pores and cavities formed by the framework structure and typical for zeolitic materials in general.

According to the present invention, a zeolitic material having a CHA framework structure is crystallized in (2), wherein said material comprises $YO_2$. Furthermore, one or more sources for $YO_2$ and seed crystals comprising $YO_2$ are provided in (1) for the crystallization thereof. In principle, Y stands for any conceivable tetravalent element, Y standing for either or several tetravalent elements. Preferred tetravalent elements according to the present invention include Si, Sn, Ti, Zr, and Ge, and combinations thereof. More preferably, Y stands for Si, Ti, or Zr, or any combination of said tetravalent elements, even more preferably for Si, and/or Sn. According to the present invention, it is particularly preferred that Y stands for Si.

Therefore, according to the inventive process, it is preferred that Y comprised in the seed crystals and/or, preferably and, that Y provided in (1) in the one or more sources for $YO_2$ are, independently from one another, selected from the group consisting of Si, Sn, Ti, Zr, Ge, and mixtures of two or more thereof, Y preferably being Si.

According to the inventive process, one or more sources for $YO_2$ are provided in (1), wherein said one or more sources may be provided in any conceivable form provided that a zeolitic material having a CHA framework structure comprising $YO_2$ and $X_2O_3$ can be crystallized in (2). Preferably, $YO_2$ is provided as such and/or as a compound which comprises $YO_2$ as a chemical moiety and/or as a compound which (partly or entirely) is chemically transformed to $YO_2$ during the inventive process. In preferred embodiments of the present invention, wherein Y stands for

Si or for a combination of Si with one or more further tetravalent elements, the source for $SiO_2$ preferably provided in (1) can also be any conceivable source. Thus, by way of example, any type of silicas and/or silicates may be used, wherein preferably the one or more sources for $YO_2$ comprises one or more silicates. As said one or more silicates, any conceivable silicate or a combination of silicates may be used wherein it is preferred that said one or more silicates comprise one or more alkaline metal silicates, the alkaline metal being preferably selected from the group consisting of Li, Na, K, and combinations of two or more thereof. According to particularly preferred embodiments, the one or more silicates comprise one or more sodium and/or one or more potassium silicates, wherein even more preferably the one or more silicates comprise one or more sodium silicates.

Within the meaning of the present invention, the term "silicate" as a preferred source for $YO_2$ generally refers to any conceivable silicates, provided that an organotemplate-free zeolitic material having a CHA framework structure may be crystallized in (2) of the inventive process. According to a particularly preferred meaning of the present invention, the term "silicate" refers to the $[SiO_3]^{2-}$ anion comprised in the particularly preferrred silicate compounds comprised in the one or more sources for $YO_2$.

Thus, according to the present invention it is preferred that the one or more sources for $YO_2$ comprises one or more silicates, preferably one or more alkali metal silicates, wherein the alkali metal is preferably selected from the group consisting of Li, Na, K, and combinations of two or more thereof, wherein more preferably the alkali metal is Na and/or K, and wherein even more preferably the alkali metal is Na.

According to particularly preferred embodiments of the present invention wherein the one or more sources for $YO_2$ provided in (1) comprises one or more silicates, it is further preferred that said one or more silicates comprise water glass. In principle, any type of water glass or combinations thereof may be used in the present invention as the one or more sources for $YO_2$ provided that a zeolitic material having a CHA framework structure may be obtained, wherein preferably sodium and/or potassium silicate is employed as water glass, more preferably sodium silicate.

Therefore, according to the present invention it is preferred that the mixture prepared in (1) comprises water glass, preferably sodium and/or potassium silicate, and even more preferably sodium silicate.

According to said preferred embodiments of the present invention wherein the one or more sources for $YO_2$ comprises one or more silicates, it is further preferred that one or more silicas are added to the mixture prepared in (1) in addition to the one or more silicates. In principle, any conceivable source of silica may be employed, provided that a zeolitic material having the CHA-type framework structure may be crystallized in (2). Thus, by way of example, any type of silica may be employed such as fumed silica, silica hydrosols, reactive amorphous silicas, silica gel, silicic acid, colloidal silica, pyrogenic silica, silicic acid esters, tetraalkoxy silanes, or mixtures of at least two of these compounds, wherein more preferably one or more silica hydrosols and/or one or more colloidal silicas are used, and even more preferably one or more colloidal silicas.

Thus, according to the present invention it is further preferred that the mixture prepared in (1) of the inventive process further comprises one or more silicas in addition to the one or more silicates, preferably one or more silica hydrosols and/or one or more colloidal silicas, and even more preferably one or more colloidal silicas in addition to the one or more silicates.

According to the present invention, $X_2O_3$ is comprised in the zeolitic material having a CHA framework structure which is crystallized in (2). Furthermore, one or more sources for $X_2O_3$ and seed crystals comprising $X_2O_3$ are provided in (1) for the crystallization thereof. In principle, X stands for any conceivable trivalent element, X standing for either one or several trivalent elements. Preferred trivalent elements according to the present invention include Al, B, In, and Ga, and combinations thereof. More preferably, X stands for Al, B, or In, or any combination of said trivalent elements, even more preferably for Al and/or B. According to the present invention, it is particularly preferred that X stands for Al.

Therefore, it is preferred according to the inventive process that X comprised in the seed crystals and/or, preferably and, that X provided in (1) in the one or more sources for $X_2O_3$ are, independently from one another, selected from the group consisting of Al, B, In, Ga, and mixtures of two or more thereof, X preferably being Al and/or B, and more preferably being Al.

In the process of the present invention, the one or more sources for $X_2O_3$ provided in (1) may be provided in any conceivable form, provided that a zeolitic material having a CHA framework structure is crystallized in (2). Preferably, $X_2O_3$ is provided as such and/or as a compound which comprises $X_2O_3$ as a chemical moiety and/or as a compound which (partly or entirely) is chemically transformed to $X_2O_3$ during the inventive process.

In more preferred embodiments of the present invention, wherein X stands for Al or for a combination of Al with one or more further trivalent elements, the source for $Al_2O_3$ provided in (1) can be any conceivable source. There can be used for example any type of alumina and aluminates, aluminum salts such as, for example, alkali metal aluminates, aluminum alcoholates, such as, for example, aluminum triisopropylate, or hydrated alumina such as, for example, alumina trihydrate, or mixtures thereof. Preferably, the source for $Al_2O_3$ comprises at least one compound selected from the group consisting of alumina and aluminates, preferably aluminates, more preferably alkali metal aluminates, wherein even more preferably, the alkali metal of the aluminate comprises one or more of the alkali metals M. Among the preferred alkali metal aluminates, the at least one source preferably comprises sodium and/or potassium aluminate, more preferably sodium aluminate. In particularly preferred embodiments of the present invention, the source for $Al_2O_3$ is sodium aluminate.

Thus, according to the inventive process it is preferred that the one or more sources for $X_2O_3$ comprises one or more aluminate salts, preferably one or more alkali metal aluminates, wherein the alkali metal is preferably selected from the group consisting of Li, Na, K, and combinations of two or more thereof, wherein more preferably the alkali metal is Na and/or K, and wherein even more preferably the alkali metal is Na.

According to particularly preferred embodiments of the inventive process, the mixture according to (1) comprises one or more silicates as a source for $YO_2$ and one or more aluminates as a source for $X_2O_3$, more preferably one or more alkali metal silicates and one or more alkali metal aluminates, more preferably a sodium and/or potassium silicate and a sodium and/or potassium aluminate, more preferably a sodium silicate and sodium aluminate, wherein even more preferably the one or more sources for $YO_2$ comprises sodium waterglass ($Na_2SiO_3$) and the one or more sources for $X_2O_3$ comprises sodium aluminate. According to alternatively particularly preferred embodiments of the present invention, the mixture according to (1) comprises one or more silicas in addition to one or more silicates as a source for $YO_2$ and one or more aluminates as a source for $X_2O_3$, more preferably one or more colloidal silicas in addition to one or more alkali metal silicates and one or more alkali metal aluminates, more preferably one or more colloidal silicas in addition to a sodium and/or a potassium silicate and sodium and/or potassium aluminate, more preferably one or more colloidal silicas in addition to a sodium silicate and sodium aluminate, wherein even more preferably the one or more sources for $YO_2$ comprises one or more colloidal silicas in addition to sodium waterglass ($Na_2SiO_3$) and the one or more sources for $X_2O_3$ comprises sodium aluminate.

According to the inventive process, the mixture prepared in (1) optionally comprises one or more sources for $Z_2O_5$, wherein Z stands for any conceivable pentavalent element, Z standing for either one or several pentavalent elements. Preferably, $Z_2O_5$ is provided as such and/or as a compound which comprises $Z_2O_5$ as a chemical moiety and/or as a compound which (partly or entirely) is chemically transformed to $Z_2O_5$ during the inventive process. In principle, any conceivable source may be provided as the one or more sources for $Z_2O_5$, provided that in (2) of the inventive process, a zeolitic material having a CHA framework structure is crystallized. Furthermore, the zeolitic material having a CHA framework structure crystallized in (2) as well as the seed crystals provided in (1) for the crystallization thereof may respectively comprise $Z_2O_5$. Preferred pentavalent elements Z according to the present invention include P, As, Sb, Bi, V, Nb, Ta, and combinations of two or more thereof. More preferably, Z stands for P, As, V, and combinations of two or more thereof, wherein even more preferably Z comprises P or As. According to particularly preferred embodiments, Z comprises P, wherein it is particularly preferred that Z stands for P.

Therefore, it is preferred according to the inventive process that Z optionally comprised in the seed crystals and/or, preferably and, that Z in the one or more sources for $Z_2O_5$ optionally further provided in (1) are, independently from one another, selected from the group consisting of P, As, Sb, Bi, V, Nb, Ta, and combinations of two or more thereof, preferably from the group consisting of P, As, V, and combinations of two or more thereof, wherein more preferably Z comprises P or As, preferably P, and wherein even more preferably Z is P.

In the process of the present invention, one or more sources for $Z_2O_5$ may optionally be provided in (1). As regards the one or more sources for $Z_2O_5$ which may be provided to this effect, no particular restrictions apply provided that a zeolitic material having a CHA framework structure is crystallized in (2). Preferably, the one or more sources for $Z_2O_5$ is provided as such and/or as a compound which comprises $Z_2O_5$ as a chemical moiety and/or as a compound which (partly or entirely) is chemically transformed to $Z_2O_5$ during the inventive process.

In more preferred embodiments of the present invention, wherein Z stands for P or for a combination of P with one or more further pentavalent elements, the source for $P_2O_5$ provided in (1) can be any conceivable source. There can be used for example one or more phosphates and/or one or more oxides and/or one or more acids of phosphorous, wherein preferably one or more acids of phosphorous is employed, and more preferably phosphoric acid. According to the present invention it is particularly preferred that phosphoric acid is employed that the source for $Z_2O_5$ optionally provided in step (1).

According to the inventive process, the mixture prepared in (1) comprises seed crystals having a CHA framework structure, wherein the CHA framework structure of the seed crystals comprises $YO_2$ and $X_2O_3$, wherein if the CHA framework structure of the seed crystals does not contain $Z_2O_5$, said seed crystals then preferably display a $YO_2 : X_2O_3$ molar ratio of 5 or greater than 5. Thus, it has been found that by using seed crystals having a CHA framework structure comprising $YO_2$ and $X_2O_3$, wherein the $YO_2 : X_2O_3$ molar ratio is 5 or greater than 5, an organotemplate-free synthetic process may be provided for obtaining a zeolitic material having a CHA framework structure comprising $YO_2$ and $X_2O_3$, in particular in instances in which the CHA framework structure of the seed crystals does not contain $Z_2O_5$. In particular, the process of the present invention preferably allows for the use of seed crystals displaying intermediate and high $YO_2 : X_2O_3$ molar ratios. Within the meaning of the present invention, intermediate and high $YO_2 : X_2O_3$ molar ratios generally refers to molar ratios having a value of 5 or greater than 5, and in particular to $YO_2 : X_2O_3$ molar ratios according to particular and preferred embodiments of the present invention. Thus, according to the present invention, the $YO_2 : X_2O_3$ molar ratio of the seed crystals having a CHA framework structure provided in (1) of the inventive process may display any conceivable $YO_2 : X_2O_3$ molar ratio provided that a zeolitic material having a CHA framework structure may be crystallized in (2) of the inventive process, wherein the value of the $YO_2 : X_2O_3$ molar ratio is preferably either 5 or a value greater than 5. Thus, by way of example, the seed crystals provided in (1) of the inventive process may display a $YO_2 : X_2O_3$ molar ratio in the range of anywhere from 5 to 200 wherein it is preferred that the molar ratio displayed by the seed crystals is comprised in the range of from 6 to 150, more preferably of from 8 to 100, more preferably of from 12 to 70, more preferably of from 20 to 50, more preferably of from 25 to 40, and even more preferably of from 28 to 35. According to the inventive process it is particularly preferred that the $YO_2 : X_2O_3$ molar ratio of the seed crystals is in the range of from 29 to 33.

Therefore, it is preferred according to the inventive process that optionally one or more sources for $Z_2O_5$ are further provided in (1), and that if the CHA framework structure of the seed crystals does not contain $Z_2O_5$, the seed crystals then have a $YO_2 : X_2O_3$ molar ratio of 5 or greater than 5.

According to the present invention, the seed crystals having a CHA framework structure provided in (1) of the inventive process may optionally comprise $Z_2O_5$. In said instances, the seed crystals having a CHA framework structure provided in (1) of the inventive process display a $YO_2 : nX_2O_3 : pZ_2O_5$ molar ratio More specifically, according to said optional embodiments of the present invention wherein the CHA framework structure of the seed crystals further comprises $Z_2O_5$, said crystals are not necessarily characterized by a $YO_2 : X_2O_3$ molar ratio but rather by a $YO_2 : nX_2O_3 : pZ_2O_5$ molar ratio wherein the value for the ratio $(1+2p) : (n-p)$ is characteristic for the CHA framework structure of said seed crystals.

Thus, according to said embodiments of the present invention wherein the CHA framework structure of the seed crystals further comprises $Z_2O_5$, there is no particular restriction as to the $YO_2 : X_2O_3$ molar ratio which the seed crystals further comprising $Z_2O_5$ may display. Thus, considering the value for the ratio $(1+2p) : (n-p)$, said ratio may have any suitable value, provided that an organotemplate-free zeolitic material may be crystallized in (2). Thus by way of example, the value for the ratio $(1+2p) : (n-p)$ may be 1 or greater than 1, wherein it is preferred that said value is 2 or greater than 2, more preferably 3 or greater than 3, and even more preferably 5 or greater than 5. Thus, according to preferred embodiments of the inventive process, the value for the ratio $(1+2p) : (n-p)$ of seed crystals further comprising $Z_2O_5$ may range anywhere from 1 to 500, more preferably from 2 to 400, more preferably from 3 to 300, more preferably from 5 to 200, more preferably from 6 to 150, more preferably from 8 to 100, more preferably from 12 to 70, more preferably from 20 to 50, more preferably from 25 to 40, and even more preferably from 28 to 35. and even more preferably from 29 to 33. According to particularly preferred optional embodiments of the present invention, the value for the ratio $(1+2p) : (n-p)$ relative to the $YO_2 : nX_2O_3 : pZ_2O_5$ molar ratio of the seed crystals is comprised in the range of from 29 to 33.

Thus, it is preferred according to the inventive process that if the CHA framework structure of the seed crystals further comprises $Z_2O_5$ in addition to $YO_2$ and $X_2O_3$, the seed crystals then have a $YO_2 : nX_2O_3 : pZ_2O_5$ molar ratio, wherein the value for the ratio $(1+2p) : (n-p)$ is 5 or greater than 5, wherein the value for the ratio $(1+2p) : (n-p)$ preferably ranges from 5 to 200, more preferably from 6 to 150, more preferably from 8 to 100, more preferably from 12 to 70, more preferably from 20 to 50, more preferably from 25 to 40, more preferably from 28 to 35, and even more preferably from 29 to 33.

Regarding the composition of the seed crystals having a CHA framework structure, there is no particular restriction as to their composition, provided that they comprise $YO_2$ and $X_2O_3$, wherein Y is a tetravalent element and X is a trivalent element, and provided that they are suitable for crystallizing an organotemplate-free zeolitic material having a CHA framework structure in (2) of the inventive process. As defined in the foregoing with respect to the inventive process, $YO_2$ and $X_2O_3$ comprised in the framework structure of the seed crystals having a CHA framework structure is contained therein as structure building elements, as opposed to non-framework elements which can be present in the pores and/or cavities formed by the CHA framework structure and typical for zeolitic materials in general.

In principle, the seed crystals having a CHA framework structure may comprise any conceivable tetravalent element Y, wherein Y stands for one or several tetravalent elements. Preferred tetravalent elements comprised in the seed crystals according to the present invention include Si, Sn, Ti, Zr, and Ge, and combinations of two or more thereof. More preferably, Y stands for Si, Ti, or Zr, or any combination of said tetravalent elements, even more preferably for Si and/or Sn. According to the present invention, it is particularly preferred that Y stands for Si.

According to the present invention it is further preferred that both said one or more sources for $YO_2$ and the seed crystals having a CHA framework structure provided in (1) comprise the same one or more tetravalent elements, wherein even more preferably Y comprised in the one or more sources for $YO_2$ and Y comprised in the seed crystals having a CHA framework structure stand for the same one or more tetravalent elements according to particular and preferred embodiments of the present invention.

Furthermore, the seed crystals having a CHA framework structure may comprise any suitable trivalent element X, wherein again X stands for either one or several trivalent elements. Preferred trivalent elements according to the present invention include Al, B, In, and Ga, and combinations thereof. More preferably, X comprises Al or Ga, wherein more preferably X comprises Al, and wherein even more preferably X is Al.

As for the one or more tetravalent elements Y, it is further preferred according to the inventive process that both said one or more sources for $X_2O_3$ and the seed crystals having a CHA framework structure provided in (1) comprise the same one or more trivalent elements X, wherein even more preferably X comprised in the one or more sources for $X_2O_3$ and X comprised in the seed crystals having a CHA framework structure stand for the same one or more trivalent elements according to particular and preferred embodiments of the present invention.

Finally, as regards the use of seed crystals optionally comprising $Z_2O_5$ in the inventive process, the CHA framework structure may accordingly comprise any suitable pentavalent element Z, wherein Z stands for either one of several pentavalent elements. Preferred pentavalent elements according to the present invention include P, As, Sb, Bi, V, Nb, Ta, and combinations of two or more thereof. More preferably Z comprises one or more pentavalent elements selected from the group consisting of P, As, V, and combinations of two or more thereof, wherein more preferably Z comprises P or As, preferably P, and wherein even more preferably Z is P

According to particularly preferred embodiments, both said one or more sources for $YO_2$ and said one or more sources for $X_2O_3$ as well as the seed crystals having a CHA framework structure provided in (1) comprise the same one or more tetravalent elements Y in addition to the same one or more trivalent elements X, wherein even more preferably Y comprised in the one or more sources for $YO_2$ and Y comprised in the seed crystals as well as X comprised in the one or more sources for $X_2O_3$ and X comprised in the seed crystals stand for the same one or more tetravalent elements and the same one or more trivalent elements, respectively, according to particular and preferred embodiments of the present invention.

According to the present invention, there is no particular restriction with respect to the amount of the one or more sources for $YO_2$ and the one or more sources for $X_2O_3$ provided in the mixture in (1) of the inventive process provided that a zeolitic material having a CHA framework structure comprising $YO_2$ and $X_2O_3$ may be crystallized in (2). Thus, in principle, any conceivable $YO_2 : X_2O_3$ molar ratio may be provided in the mixture prepared in (1), wherein, by way of example, said molar ratio may range anywhere from 1 to 150. According to the present invention it is however preferred that the $YO_2 : X_2O_3$ molar ratio of the mixture prepared in (1) is comprised in the range of from 2 to 100, more preferably from 5 to 70, more preferably from 10 to 50, more preferably from 13 to 30, and more preferably from 16 to 25. According to particularly preferred embodiments of the present invention, the $YO_2 : X_2O_3$ molar ratio of the mixture prepared in (1) ranges from 18 to 22.

In the process of the present invention, the mixture prepared in (1) preferably comprises one or more alkaline metals M. In general, said one or more alkaline metals M may be provided from any suitable compounds or compounds comprising one or more alkaline metals M, wherein preferably the one or more alkaline metals M are provided as one or more alkaline metal salts. According to particularly preferred embodiments thereof, the one or more alkaline metals M are provided as one or more alkaline metal compounds being the one or more sources for $YO_2$ and/or the one or more sources for $X_2O_3$, even more preferably as one or more alkaline metal compounds being the respective sources for $YO_2$ and $X_2O_3$. According to said particularly and preferred embodiments, the one or more alkaline metal compounds used as sources for $YO_2$ and/or $X_2O_3$ preferably comprise one or more alkaline metal salts used as the one or more sources for $YO_2$ and/or $X_2O_3$ wherein according to a particularly preferred embodiment one or more alkaline metal salts are used as the one or more sources for $YO_2$ and/or $X_2O_3$, and preferably as the one or more sources for both $YO_2$ and $X_2O_3$.

As regards the one or more alkaline metals M preferably comprised in the mixture prepared in (1) of the inventive process, any suitable alkaline metal M or combination of alkaline metals M may be used, wherein preferably the one or more alkaline metals M are selected from the group consisting of Li, Na, K, and combinations of two or more thereof. According to particularly preferred embodiments, the one or more alkaline metals M comprise Li and/or Na, and preferably Na, wherein even more preferably the one or more alkaline metal is Li and/or Na, and preferably Na.

As regards the preferred embodiments of the inventive process wherein one or more alkaline metals M are provided in (1), there is no particular restriction according to the present invention as to the amount of the one or more alkaline metals M which may be provided therein, provided that a zeolitic material having a CHA framework structure is crystallized in (2). Thus, by way of example, the $M_2O : YO_2$ molar ratio of the mixture prepared in (1) according to said preferred embodiments may range anywhere from 0.01 to 5, wherein preferably said ratio ranges from 0.05 to 2, more preferably from 0.1 to 1.5, more preferably from 0.15 to 1, and even more preferably from 0.2 to 0.5. According to particularly preferred embodiments thereof, the $M_2O : YO_2$ molar ratio of the mixture prepared in (1) ranges from 0.25 to 0. 35.

Furthermore, according to preferred embodiments of the inventive process wherein the mixture prepared in (1) comprises one or more alkaline metals M, there is also no particular restriction with respect to the $M_2O : X_2O_3$ molar ratio of the mixture, again provided that a zeolitic material having a CHA framework structure is crystallized in (2). Same applies accordingly to the $YO_2 : X_2O_3 : M_2O_3$ molar ratios of said preferred mixtures comprising one or more alkaline metals M. Thus, by way of example, the $YO_2 : X_2O_3 : M_2O_3$ molar ratios of said preferred mixtures may range anywhere from (5 - 70) : 1 : (0.5 - 20), wherein preferably the molar ratios are in the range of from (10 - 50) : 1 : (1 - 15), more preferably from (13 - 30) : 1 : (2 - 10), and more preferably from (16 - 25) : 1 : (4 - 8). According to particularly preferred embodiments, the $YO_2 : X_2O_3 : M_2O_3$ molar ratios of the mixture prepared in (1) range from (18 - 22) : 1 : (5 - 7).

According to alternatively preferred embodiments of the inventive process wherein the mixture prepared in (1) comprises one or more alkaline metals M, said mixture does not contain potassium and/or does not contain strontium. According to the present invention, a mixture defined as not containing potassium and/or strontium relates to a mixture wherein the amount of potassium and/or strontium contained therein is 0.001 wt.-% or less of potassium and/or strontium, preferably of 0.0005 wt.-% or less, more preferably of 0.00001 wt.-% or less, more preferably of 0.000005 wt.-% or less, and even more preferably 0.000001 wt.-% or less thereof. Said amounts of potassium and/or strontium, if at all present in the mixture prepared in (1), may also be denoted as “impurities” or “trace amounts” within the meaning of the present invention. According to particularly preferred embodiments of the present invention, the mixture prepared in (1) does not contain potassium.

Thus, according to the present invention it is preferred that the mixture prepared in (1) contains no K and/or Sr, preferably no K.

According to the present invention, there is no particular restriction as to the CHA framework structure displayed by the seed crystals provided that said seed crystals display an X-ray diffraction pattern typical of a CHA framework structure in particular with respect to the reflections and their 2θ degree positions relative to one another. Thus, in principle, provided that the diffraction pattern is typical of a CHA framework structure, there is no particular restriction neither with respect to the actual position of the reflections measured as angle 2θ, nor with respect to the intensities of the individual reflections relative to one another. According to preferred embodiments of the present invention, however, the first reflection in the X-ray diffraction pattern of the seed crystals, i.e. the reflection having the lowest angle 2θ value is the reflection having highest intensity among all measured reflections, i.e. the reflection measured at the lowest diffraction angle 2θ has an intensity of 100 %. Furthermore, when using Cu K(alpha 1) radiation, it is further preferred that the diffraction angle 2θ having an intensity of 100 % is comprised in the range of from 5 to 15 °2θ, wherein more preferably, said most intense reflection is comprised in the range of from 8 to 12 °2θ, more preferably of from 9 to 10.5 °2θ, more preferably of from 9.2 to 10 °2θ, more preferably of from 9.5 to 9.7 °2θ, and even more preferably of from 9.55 to 9.65 °2θ. According to particularly preferred embodiments of the present invention, the most intense reflection in the diffraction pattern of the seed crystals having a CHA framework structure when using Cu K(alpha 1) radiation is comprised in the range of from 9.58 to 9.62 °2θ.

According to the process of the present invention, seed crystals are provided in (1), wherein said seed crystals comprise a zeolitic material having a CHA framework structure. In general, said seed crystals can comprise any zeolitic material having a CHA framework structure provided that a zeolitic material having a CHA framework structure is crystallized in (2), wherein if the CHA framework structure of the seed crystals does not contain $Z_2O_5$, the framework structure preferably has a $YO_2 : X_2O_3$ molar ratio of 5 or greater than 5. Preferably, the zeolitic material having a CHA framework structure comprised in the seed crystals is a zeolitic material as obtained in or obtainable from (2) of the inventive process, and in particular according to any of the particular or preferred embodiments thereof described in the present application. More preferably, the zeolitic material having a CHA framework structure comprised in the seed crystals is the same as the zeolitic material having a CHA framework structure which is crystallized in (2). Particularly preferred according to the present invention are seed crystals comprising one ore more zeolites selected from the group consisting of $(Ni(deta)_2)$-UT-6, Chabazite, |Li-Na| [Al-Si-O]-CHA, DAF-5, Na-Chabazite, K-Chabazite, LZ-218, Linde D, Linde R, MeAPSO-47, Phi, SAPO-34, SAPO-47, SSZ-13, SSZ-62, UiO-21, Willhendersonite, ZK-14, ZYT-6, and combinations of two or more thereof, preferably from the group consisting of Chabazite, |Li-Na| [Al-Si-O]-CHA, Na-Chabazite, K-Chabazite, SAPO-34, SAPO-47, SSZ-13, SSZ-62, and combinations of two or more thereof, more preferably from the group consisting of Chabazite, |Li-Na| [Al-Si-O]-CHA, Na-Chabazite, SAPO-34, SSZ-13, and combinations of two or more thereof, more preferably from the group consisting of Na-Chabazite, SAPO-34, SSZ-13, and combinations of two or more thereof, wherein even more preferably the seed crystals having a CHA framework structure comprise Na-Chabazite and/or SAPO-34, and preferably Na-Chabazite. According to further preferred embodiments of the present invention, the seed crystals provided in (1) comprise one or more of the aforementioned particular and preferred zeolites, wherein said one or more zeolites have been obtained according to the inventive process.

According to the inventive process, any suitable amount of seed crystals can be provided in the mixture according to (1), provided that a zeolitic material having a CHA framework structure is crystallized in (2). Thus, by way of example, an amount of seed crystals may be provided in the mixture according to (1) ranging anywhere from 0.1 to 35 wt.-% based on 100 wt.-% of $YO_2$ in the one or more sources for $YO_2$ provided in (1) for obtaining a zeolitic material with a CHA framework structure in (2). According to the present invention it is however preferred that an amount of from 0.5 to 28 wt.-%, more preferably from 1 to 22 wt.-%, more preferably from 5 to 18 wt.-%, more preferably from 10 to 15 wt.-%. According to a particularly preferred embodiment of the present invention, the amount of seed crystals in the mixture according to (1) ranges from 12 to 14 wt.-% based on 100 wt.-% of $YO_2$ in the one ore more sources for $YO_2$.

According to the present invention, the mixture according to (1) of the inventive process preferably further comprises one or more solvents. In this respect, any conceivable solvents may be used in any suitable amount, provided that a zeolitic material having a CHA framework structure comprising $YO_2$, $X_2O_3$, and optionally comprising $Z_2O_5$ can be obtained from crystallization in (2). Thus, by way of example, the one or more solvents may be chosen from water, organic solvents, and mixtures thereof, preferably from the group consisting of distilled water, alcohols, and mixtures thereof. More preferably from the group consisting of distilled water, methanol, ethanol, propanol, and mixtures thereof. According to particularly preferred embodiments of the present invention, only water and preferably only distilled water is contained in the mixture according to (1) as the solvent.

Therefore, it is preferred according to the inventive process, that the mixture prepared in (1) further comprises one or more solvents, wherein said one or more solvents preferably comprises water, more preferably distilled water.

As regards the one or more solvents which are preferably provided in the mixture according to (1), any suitable amount thereof can be used. Thus, with respect to preferred embodiments wherein water and preferably distilled water is comprised in the mixture according to (1) as a solvent, and wherein even more preferably only distilled water is contained in the mixture, the $H_2O : YO_2$ molar ratio of the mixture prepared in (1) may range anywhere from 1 to 25. According to said preferred embodiments, however, it is preferred that the $H_2O : YO_2$ molar ratio ranges from 2 to 20, more preferably from 4 to 18, more preferably from 6 to 16, and more preferably from 8 to 14. According to particularly preferred embodiments of the present invention, wherein the mixture according to (1) comprises water as a solvent, and even more preferably wherein distilled water is the only solvent present in said mixture, it is preferred that the $H_2O : YO_2$ molar ratio of the mixture according to (1) ranges from 10 to 12.

In general, (2) according to the inventive process can be conducted in any conceivable manner, provided that a zeolitic material having a CHA framework structure is crystallized from the mixture according to (1). The mixture can be crystallized in any type of vessel, wherein a means of agitation is preferably employed, preferably by rotation of the vessel and/or stirring, and more preferably by stirring the mixture.

According to the inventive process, the mixture is preferably heated during at least a portion of the crystallization process in (2). In general, the mixture can be heated to any conceivable temperature of crystallization, provided that a zeolitic material having a CHA framework structure is crystallized from the mixture. Preferably, the mixture is heated in (2) to a temperature of crystallization ranging from 80 to 200°C, more preferably from 90 to 180°C, more preferably from 100 to 160°C, more preferably from 110 to 140°C, and even more preferably from 115 to 130°C.

In preferred embodiments of the present invention, the mixture according to (1) is subjected in (2) to a pressure which is elevated with regard to normal pressure. The term "normal pressure" as used in the context of the present invention relates to a pressure of 101,325 Pa in the ideal case. However, this pressure may vary within boundaries known to the person skilled in the art. By way of example, this pressure can be in the range of from 95,000 to 106,000 or of from 96,000 to 105,000 or of from 97,000 to 104,000 or of from 98,000 to 103,000 or of from 99,000 to 102,000 Pa.

In preferred embodiments of the inventive process wherein a solvent is present in the mixture according to (1), it is furthermore preferred that heating in (2) is conducted under solvothermal conditions, meaning that the mixture is crystallized under autogenous pressure of the solvent which is used, for example by conducting heating in an autoclave or other crystallization vessel suited for generating solvothermal conditions. In particularly preferred embodiments wherein the solvent comprises water, preferably distilled water, heating in (2) is accordingly preferably conducted under hydrothermal conditions.

The apparatus which can be used in the present invention for crystallization is not particularly restricted, provided that the desired parameters for the crystallization process can be realized, in particular with respect to the preferred embodiments requiring particular crystallization conditions. In the preferred embodiments conducted under solvothermal conditions, any type of autoclave or digestion vessel can be used.

According to particular and preferred embodiments of the present invention, wherein the mixture is heated in (2), said heating may be conducted during the entire crystallization process or during only one or more portions thereof, provided that a zeolitic material having the CHA framework structure is crystallized. Preferably, heating is conducted during the entire duration of crystallization.

In general, the duration of the crystallization process in (2) of the inventive process is not particularly limited. In preferred embodiments involving heating of the mixture according to (1), said crystallization process is conducted for a period ranging from 2 to 36 h, more preferably from 6 to 24 h, more preferably from 12 to 20 h, and more preferably from 14 to 18 h.

In general, the process of the present invention can optionally comprise further steps for the work-up and/or further physical and/or chemical transformation of the zeolitic material having an CHA framework structure crystallized in (2) from the mixture prepared in (1). The crystallized material can for example be subject to any sequence of isolation and/or washing procedures, wherein the zeolitic material obtained from crystallization in (2) is preferably subject to at least one isolation and at least one washing procedure.

Therefore, the inventive process preferably further comprises one or more of the following
(2a) isolating the zeolitic material having a CHA framework structure, preferably by filtration,
and/or
(2b) washing the zeolitic material having a CHA framework structure,
and/or
(2c) drying the zeolitic material having a CHA framework structure,
wherein the steps (2a) and/or (2b) and/or (2c) can be conducted in any order, and
wherein one or more of said steps is preferably repeated one or more times.

Isolation of the crystallized product in (2a) can be achieved by any conceivable means. Preferably, isolation of the crystallized product in (2a) is achieved by means of filtration, ultrafiltration, diafiltration, centrifugation and/or decantation methods, wherein filtration methods can involve suction and/or pressure filtration steps.

With respect to one or more preferred washing procedures in (2b), any conceivable solvent can be used. Washing agents which may be used are, for example, water, alcohols, such as methanol, ethanol or propanol, or mixtures of two or more thereof. Examples of mixtures are mixtures of two or more alcohols, such as methanol and ethanol or methanol and propanol or ethanol and propanol or methanol and ethanol and propanol, or mixtures of water and at least one alcohol, such as water and methanol or water and ethanol or water and propanol or water and methanol and ethanol or water and methanol and propanol or water and ethanol and propanol or water and methanol and ethanol and propanol. Water or a mixture of water and at least one alcohol, preferably water and ethanol, is preferred, distilled water being very particularly preferred as the only washing agent in (2b).

Preferably, the separated zeolitic material is washed in (2b) until the pH of the washing agent, preferably the washwater, is in the range of from 6 to 8, preferably from 6.5 to 7.5, as determined via a standard glass electrode.

In general, any conceivable means of drying can be used in (2c). Drying procedures preferably include heating and/or applying vacuum to the zeolitic material having a CHA framework structure. According to the present invention, it is particularly preferred that the one or more drying steps in (2c) involve spray drying and/or spray granulation, and preferably spray drying of the zeolitic material.

In particular and preferred embodiments of the inventive process comprising at least one drying step (2c), the drying temperatures are preferably in the range of from 25°C to 150°C, more preferably of from 60 to 140°C, more preferably of from 70 to 130°C and even more preferably in the range of from 75 to 125°C. The durations of drying are preferably in the range of from 2 to 60 h, more preferably in the range of 6 to 48 hours, more preferably of from 12 to 36 h, and even more preferably of from 18 to 30 h.

According to the inventive process, the zeolitic material crystallized in (2) is subject to at least one ion-exchange procedure in (3), wherein the term "ion-exchange" according to the present invention generally refers to non-framework ionic elements and/or molecules contained in the zeolitic material. Preferably, the non-framework ionic element comprises one or more of the one or more alkali metals M preferably comprised in the zeolitic material having a CHA framework structure, more preferably Na and/or K, and even more preferably Na which is accordingly ion-exchanged against against Cu and/or Fe, and preferably against Cu. In general, any conceivable ion-exchange procedure with all possible ionic elements and/or molecules can be conducted on the zeolitic material, with the exception of organic structure directing agents specifically used in the synthesis of zeolitic materials having an CHA framework structure, in particular specific tetraalkylammonium compounds, dialkyl amines, heterocyclic amines, including combinations of two or more thereof, and/or related organotemplates such as any suitable N-alkyl-3-quinuclidinol compound, N,N,N-trialkyl-exoaminonorbornane compound, N,N,N-trimethyl-1-adamantylammonium compound, N,N,N-trimethyl-2-adamantylammonium compound, N,N,N-trimethylcyclohexylammonium compound, N,N-dimethyl-3,3-dimethylpiperidinium compound, N,N-methylethyl-3,3-dimethylpiperidinium compound, N,N-dimethyl-2-methylpiperidinium compound, 1,3,3,6,6-pentamethyl-6-azonio-bicyclo(3.2.1)octane compound, N,N-dimethylcyclohexylamine compound, or any suitable N,N,N-trimethylbenzylammonium compound, including combinations of two or more thereof.

Therefore, it is preferred according to the inventive process that in (3) at least one ionic non-framework element contained in the zeolitic material having a CHA framework is ion exchanged against Cu and/or Fe, and preferably against Cu, wherein said at least one ionic non-framework element is preferably one or more alkali metals, more preferably Na and/or K, and more preferably Na.

According to the inventive process, the zeolitic material obtained in (2) is subject to an ion-exchange procedure, wherein it is ion-exchanged against copper and/or iron, preferably against copper. In general, any conceivable ion-exchange procedure can be conducted on the zeolitic material to this effect, provided that a copper and/or iron ion-exchanged zeolitic material is obtained. It is, however, preferred according to the present invention that the zeolitic material obtained in (2) is first converted to the H-form, preferably via the ammonium form and subsequent calcination thereof for obtaining the H-form, prior to ion change with copper and/or iron.

Therefore, it is preferred according to the inventive process that (3) comprises one or more of the following

(3a) subjecting the zeolitic material obtained in (2) to one or more ion exchange procedures with $H^+$ and/or $NH_4^+$, preferably with $NH_4^+$;

(3b) optionally calcining the zeolitic material obtained in (3a); and

(3c) subjecting the zeolitic material obtained in (3b) to one or more ion exchange procedures with Cu and/or Fe.

As regards the amount of copper and/or iron is ion exchanged into the zeolitic material according to the inventive process, no particular restrictions apply provided that the amount of copper and/or iron may exchanged therein is in the range of from from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $YO_2$ contained in the zeolitic material having a CHA framework structure. Within the meaning of the present invention, reference to the amounts of copper and/or iron as such or in relation to other materials indicates the total amount of copper and/or iron or refers to the totality of copper and/or iron contained in the zeolitic material. Thus, with respect to materials obtained according to the inventive process as well as to inventive materials containing both copper and iron, respectively, the term "copper and/or iron" as used in the present application accordingly indicates the total amount or the totality of both copper and iron contained in the inventive zeolitic material or in the zeolitic material obtained according to the inventive process. Preferably, the zeolitic material is ion exchanged in (3) such as to obtain a loading in the zeolitic material ranging from 4.5 to 10 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $YO_2$ contained in the zeolitic material having a CHA framework structure. It is, however, preferred according to the inventive process that the zeolitic material is ion exchanged such as to obtain a loading of copper and/or iron ranging from 5.2 to 9.5 wt.-%, more preferably from 5.5 to 9 wt.-%, more preferably from 6 to 8.5 wt.-%, and more preferably from 6.5 to 8 wt.-%. According to the inventive process it is particularly preferred that the zeolitic material is ion exchanged in (3) such as to obtain a loading of copper and/or iron ranging from 7 to 7.5 wt.-% calculated as the respective element and based on 100 wt-% of $YO_2$ contained in the zeolitic material having a CHA framework structure.

Concerning the state in which copper and/or iron is ion exchanged into the zeolitic material, it is noted that no particular restrictions apply according to the present invention. Thus, in principle, copper and iron may be ion exchanged as $Cu^+$, $Cu^{2+}$, $Fe^{2+}$, and/or $Fe^{3+}$, respectively, wherein it is however preferred according to the present invention that independently from one another copper is ion exchanged as $Cu^{2+}$ and iron is ion exchanged as $Fe^{2+}$.

As outlined in the forgoing, the ion-exchange step (3) may comprise one or more ion-exchange procedures. According to (3a), the zeolitic material obtained in step (2) of the inventive process is first ion-exchanged with $H^+$ and/or ammonium, preferably with $H^+$, prior to one or more ion-exchange procedures with copper and/or iron. According to the present invention, the zeolitic material obtained in step (3a) of the inventive process may also be subject to a step of calcination in (3b), is effected by heating of the zeolitic material, prior to ion-exchange with copper and/or iron in step (3c). According to said optional embodiments of the present invention, wherein the zeolitic material obtained in step (3a) is calcined, said calcination may be effected in (3b) by heating of the zeolitic material to any suitable temperature for any conceivable duration provided that the resulting material may be ion-exchanged with copper and/or iron for obtaining an ion-exchanged material wherein the loading with copper and/or iron is comprised in the range of from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $YO_2$ contained in the zeolitic material having a CHA framework structure. Thus, by way of example, the calcination temperature may range anywhere from 200 to 850°C, wherein the calcination temperature is preferably comprised in the range of from 250 to 750°C, more preferably from 300 to 700°C, more preferably from 350 to 650 °C, more preferably from 400 to 600°C, and more preferably from 450 to 550°C. According to particularly preferred embodiments of the present invention including a calcination step prior to ion-exchange in (3c), the zeolitic material obtained in step (3a) is calcined in (3b) at a temperature comprised in the range of from 475 to 525°C prior to ion-exchange with copper and/or iron in (3c). Furthermore, as regards the duration of the calcination procedure optionally used prior to ion-exchange in step (3c), the calcination may be conducted for a period ranging anywhere from 0.5 to 36 h, wherein preferably the duration of the calcination ranges from 1 to 24 h, more preferably from 2 to 20 h, more preferably from 3 to 16 h, more preferably from 3.5 to 12 h, more preferably from 4 to 10 h, more preferably from 4.5 to 8 h, more preferably from 5 to 7 h. According to particularly preferred embodiments, the calcination procedure performed to ion-exchange with copper and/or iron is performed for a duration of from 5.5 to 6.5 h.

According to the present invention it is further preferred that the zeolitic material obtained from crystallization in (3a) and/or (3c) is subject to at least one isolating step prior to being subject to at least one ion-exchange procedure, preferably to at least one isolating step followed by at least one washing step, and more preferably to at least one isolating step followed by at least one washing step followed by at least one drying step. In general, the preferred washing and/or isolation and/or ion-exchange procedures comprised in the inventive process can be conducted in any conceivably order and repeated as often as desired.

Therefore, it is further preferred according to the inventive process that (3a) further comprises one or more of the following

(3a.i) isolating the zeolitic material ion exchanged with $H^+$ and/or $NH_4^+$, preferably by filtration,

and/or

(3a.ii) washing the zeolitic material ion exchanged with $H^+$ and/or $NH_4^+$,

and/or

(3a.iii) drying the zeolitic material ion exchanged with $H^+$ and/or $NH_4^+$,

wherein the steps (3a.i) and/or (3a.ii) and/or (3a.iii) can be conducted in any order, and wherein one or more of said steps is preferably repeated one or more times.

Furthermore, and independently thereof, it is further preferred according to the inventive process that (3c) further comprises one or more of the following

(3c.i) isolating the zeolitic material ion exchanged with Cu and/or Fe, preferably by filtration,

and/or

(3c.ii) washing the zeolitic material ion exchanged with Cu and/or Fe,

and/or

(3c.iii) drying the zeolitic material ion exchanged with Cu and/or Fe,

wherein the steps (3c.i) and/or (3c.ii) and/or (3c.iii) can be conducted in any order, and wherein one or more of said steps is preferably repeated one or more times

Independently from one another, isolation of the crystallized product in (3a.i) and (3c.i) can be achieved by any conceivable means. Preferably, isolation of the crystallized product in (3a.i) and/or (3c.i) is achieved by means of filtration, ultrafiltration, diafiltration, centrifugation and/or decantation methods, wherein filtration methods can involve suction and/or pressure filtration steps.

With respect to one or more preferred washing procedures in (3a.i) and/or (3c.i), any conceivable solvent can be used. Washing agents which may be used are, for example, water, alcohols, such as methanol, ethanol or propanol, or mixtures of two or more thereof. Examples of mixtures are mixtures of two or more alcohols, such as methanol and ethanol or methanol and propanol or ethanol and propanol or methanol and ethanol and propanol, or mixtures of water and at least one alcohol, such as water and methanol or water and ethanol or water and propanol or water and methanol and ethanol or water and methanol and propanol or water and ethanol and propanol or water and methanol and ethanol and propanol. Water or a mixture of water and at least one alcohol, preferably water and ethanol, is preferred, distilled water being very particularly preferred as the only washing agent in (3a.i) and/or (3c.i).

Preferably, the separated zeolitic material is washed in (3a.ii) and/or (3c.ii) until the pH of the washing agent, preferably the washwater, is in the range of from 6 to 8, preferably from 6.5 to 7.5, as determined via a standard glass electrode.

Furthermore, any conceivable means of drying can be used in (3a.iii) and/or (3c.iii). Drying procedures preferably include heating and/or applying vacuum to the zeolitic material having a CHA framework structure. According to the present invention, it is particularly preferred that the one or more drying steps in (3a.iii) and/or (3c.iii) involve spray drying and/or spray granulation, and preferably spray drying of the zeolitic material.

In particular and preferred embodiments of the inventive process comprising at least one drying step (3a.iii) and/or (3c.iii), the drying temperatures are preferably in the range of from 25°C to 150°C, more preferably of from 60 to 140°C, more preferably of from 70 to 130°C and even more preferably in the range of from 75 to 125°C. The durations of drying are preferably in the range of from 2 to 60 h, more preferably in the range of 6 to 48 hours, more preferably of from 12 to 36 h, and even more preferably of from 18 to 30 h.

In the process of the present invention, it is preferred that a calcination step is not employed prior to the one or more ion exchange procedures of the inventive process. In general, a calcination step involves the heating of the zeolitic material crystallized according to (2) above a temperature of 500°C. More preferably, a process according to the present invention for the production of a zeolitic material having a CHA framework structure which does not comprise a calcination step prior to the one or more ion exchange procedures refers to processes, wherein the zeolitic material crystallized according to (2) is not subject in a subsequent step prior to ion exchange in (3) according to any of the particular and preferred embodiments of the inventive process to a temperature exceeding 450°C, more preferably 350°C, more preferably 300°C, more preferably 250°C, more preferably 200°C, and even more preferably 150°C. According to the present invention it is particularly preferred that after completion of (2) of the inventive process, wherein the crystallized zeolitic material is at ambient temperature, said material is subsequently not subject to any heating process normally or suitably conducted for removal of organotemplates form a zeolitic material having a CHA framework structure prior to ion exchange in (3) according to any of the particular and preferred embodiments of the inventive process. Within the meaning of the present invention, a zeolitic material which is “non-calcined” is one which has not been subject to any one of the aforementioned calcination procedures.

In general, the zeolitic material having a CHA framework structure obtained according to the inventive process may be any conceivable zeolite of the CHA framework type comprising $YO_2$, $X_2O_3$, and optionally comprising $Z_2O_5$, wherein Y is a tetravalent element, X is a trivalent element, and Z is a pentavalent element.

The present invention furthermore relates to a synthetic copper and/or iron containing zeolitic material having a CHA framework structure which is either obtained by the process according to the present invention or by any conceivable process which leads to a copper and/or iron containing zeolitic material having a CHA framework structure as obtainable according to the inventive process. According to particularly preferred embodiments, the synthetic copper and/or iron containing zeolitic material having a CHA framework structure is a non-calcined zeolitic material which is either obtained by the process according to the present invention or by any conceivable process which leads to a zeolitic material having a copper and/or iron containing CHA framework structure as obtainable according to the inventive process. Within the meaning of the present invention, a material which is designated as a “synthetic” material does not signify that the designated material as such may not naturally occur in nature. In particular, a “synthetic” material only indicates that it is manmade but by no means excludes that the material as such may occur naturally. In this respect, it is herewith further noted that within the meaning of the present invention, the term “organotemplate-free zeolitic material” is synonymous to “synthetic organotemplate-free zeolitic material”.

Therefore, the present invention also relates to a synthetic copper and/or iron containing zeolitic material having a CHA framework structure, optionally obtainable and/or obtained according to the process of any of the particular and preferred embodiments of the inventive process, wherein the CHA framework structure comprises $SiO_2$, $X_2O_3$, and optionally comprises $Z_2O_5$, wherein X is a trivalent element, and Z is a pentavalent element,
wherein the zeolitic material contains from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure, and
wherein the $^{29}$Si MAS NMR of the zeolitic material comprises:

a first peak (P1) in the range of from −96 to −98.8 ppm;
a second peak (P2) in the range of from −102 to −104.5 ppm; and
a third peak (P3) in the range of from −107.5 to −111 ppm;

wherein the integration of the first, second, and third peaks in the $^{29}$Si MAS NMR of the zeolitic material offers a ratio of the integration values P1 : P2 : P3 ranging from (0.35 – 0.7) : 1 : (0.1 – 1.6).

As regards the $^{29}$Si MAS NMR of the inventive zeolitic material, there is no particular restriction as to the number and/or respective ppm values and/or relative intensities of the signals displayed in the NMR spectrum provided that the $^{29}$Si MAS NMR comprises a first peak (P1) comprised in the range of from −96 to −98.8 ppm, a second peak (P2) comprised in the range of from −102 to −104.5 ppm, and a third peak (P3) comprised in the range of from −107.5 to −111 ppm, wherein the integration of the first, second, and third peaks in the $^{29}$Si MAS NMR of the zeolitic material offers a ratio of the integration values P1 : P2 : P3 of (0.35 – 0.7) : 1 : (0.1 – 1.6). In particular, the signal at -109 ppm of the $^{29}$Si MAS NMR corresponds to Q4 structures, wherein the respective signals at -103 and -98 ppm are attributed to the Q3 or to Q4 structures.

According to preferred embodiments, the first peak (P1) in the $^{29}$Si MAS NMR of the inventive zeolitic material is comprised in the range of from −96.5 to −98.5 ppm, more preferably of from −96.8 to −98. 2 ppm, more preferably of from −97 to −98 ppm, and more preferably of from −97.3 to −97.9 ppm. It is however particularly preferred according to the present invention that the first peak (P1) in the $^{29}$Si MAS NMR is comprised in the range of from −97.5 to −97.7 ppm. Furthermore, the second peak (P2) in the $^{29}$Si MAS NMR of the inventive zeolitic material is preferably comprised in the range of from −102.5 to −104 ppm, more preferably of from −102.8 to −103.7 ppm, and more preferably of from −103 to −103.5 ppm, wherein according to particularly preferred embodiments the second peak (P2) is comprised in the range of from −103.2 to −103.4 ppm. Finally, it is further preferred that the third peak (P3) in the $^{29}$Si MAS NMR of the inventive zeolitic material is comprised in the range of from −108 to −110.5 ppm, more preferably of from −108.5 to −110 ppm, more preferably of from −108.8 to −109.5 ppm, and more preferably of from −109 to −109.4 ppm. According to particularly preferred embodiments, however, the third peak (P3) in the $^{29}$Si MAS NMR is comprised in the range of from −109.1 to −109.3 ppm.

Furthermore, as to the integration of the first, second, and third peaks in the $^{29}Si$ MAS NMR of the zeolitic material offering a ratio of the integration values P1 : P2 : P3, according to particularly preferred embodiments, said ratio ranges from preferably from (0.4 – 0.65) : 1 : (0.15 – 1.3), more preferably from (0.42 – 0.62) : 1 : (0.2 – 1), more preferably from (0.45 – 0.6) : 1 : (0.3 – 0.7), more preferably from (0.48 – 0.58) : 1 : (0.35 – 0.5), and more preferably from (0.5 – 0.56) : 1 : (0.4 – 0.45),. According to particularly preferred embodiments of the present invention, the ratio of the integration values P1 : P2 : P3 ranges from (0.52 – 0.54) : 1 : (0.42 – 0.44).

There is no particular restriction according to the present invention as to the standard used in the $^{29}Si$ MAS NMR experiments for obtaining the respective values for the chemical shift in ppm in the $^{29}Si$ MAS NMR spectra according to particular and preferred embodiments of the present invention, wherein preferably an external standard is used. According to particularly preferred embodiments, the external standard used in the $^{29}Si$ MAS NMR experiment is the polymer Q8M8, wherein the resonance of the trimethylsilyl M group is set to 12.5 ppm. Thus, according to the present invention, the chemical shift values in ppm defined in the present application relative to the $^{29}Si$ MAS NMR spectra of the inventive zeolitic materials having a CHA framework structure are preferably based on the use of the polymer Q8M8 as an external secondary standard in the $^{29}Si$ MAS NMR experiment, wherein the resonance of the trimethylsilyl M group is set to 12.5 ppm. According to the present invention it is particularly preferred that the ppm and integration values respectively defined for the $^{29}Si$ MAS NMR of the inventive zeolitic material according to any of the particular and preferred embodiments of the present invention refers to the values obtained as described in the experimental section of the present application.

As regards the amount of copper and/or iron which is contained in the zeolitic material according to the present invention, no particular restrictions apply provided that the amount of copper and/or iron is in the range of from from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure. Preferably, the total loading of copper and/or iron in the zeolitic material ranges from 4.5 to 10 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure, more preferably from 5.2 to 9.5 wt.-%, more preferably from 5.5 to 9 wt.-%, more preferably from 6 to 8.5 wt.-%, and more preferably from 6.5 to 8 wt.-%. According to the present invention it is particularly preferred that the zeolitic material contains copper and/or iron in an amount ranging from 7 to 7.5 wt.-% calculated as the respective element and based on 100 wt-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure.

According to the present invention there is no particular restriction as to the state in which copper and/or iron is contained in the zeolitic material. It is, however, preferred according to the present invention that copper and/or iron is contained in the inventive zeolitic material at least in part and preferably entirely as extra-framework ions, wherein preferably copper is contained therein as $Cu^{2+}$, and iron is contained therein as $Fe^{2+}$ and/or $Fe^{3+}$, and preferably at least in part as $Fe^{2+}$. Furthermore, it is preferred according to the present invention that the copper and/or iron contained in the zeolitic material has at least in part been introduced therein via ion exchange. Within the meaning of the present invention, the term "extra-framework ions" designates ions which are located at the ion exchange sites of the zeolitic material and thus serve to compensate the charge of the zeolite framework structure, and in particular the negative charge thereof. Furthermore, within the meaning of the present invention, the terms "non-framework ionic elements" and "extra-framework ions" are preferably employed synonymously.

Apart from comprising $SiO_2$, the inventive copper and/or iron containing zeolitic material having a CHA framework structure comprises $X_2O_3$. In principle, X stands for any conceivable trivalent element, X standing for one or several trivalent elements. Preferred trivalent elements according to the present invention include Al, B, In, and Ga, and combinations thereof. More preferably, X stands for Al, B, or In, or any combination of said trivalent elements, even more preferably for Al and/or B. According to the present invention, it is particularly preferred that X stands for Al.

Furthermore, according to the present invention, the inventive copper and/or iron containing zeolitic material having a CHA framework structure optionally comprises $Z_2O_5$, wherein Z stands for any conceivable pentavalent element, Z standing for either one or several pentavalent elements. Preferred pentavalent elements Z according to the present invention include P, As, Sb, Bi, V, Nb, Ta, and combinations of two or more thereof. More preferably, Z stands for P, As, V, and combinations of two or more thereof, wherein even more preferably Z comprises P or As. According to particularly preferred embodiments, Z comprises P, wherein it is particularly preferred that Z stands for P

According to the present invention, there is no particular restriction as to the relative amounts of $SiO_2$ and $X_2O_3$ which may be contained in the CHA framework structure. Thus, in instances wherein the CHA framework structure does not contain $P_2O_5$, and preferably does not contain $Z_2O_5$, in an amount greater than 1 wt.-% based on 100 wt.-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure, the CHA framework structure may have an $SiO_2 : X_2O_3$ molar ratio of 7 or greater than 7. According to the present invention it is however preferred that in instances wherein the CHA framework structure does not contain $P_2O_5$, and preferably does not contain $Z_2O_5$, in an amount greater than 1 wt.-%, the CHA framework structure has an $SiO_2 : X_2O_3$ molar ratio ranging from 7.2 to 12, more preferably from 7.4 to 10, more preferably from 7.6 to 9.5, more preferably from 7.8 to 9, more preferably from 7.9 to 8.5, and more preferably from 8 to 8.2. Furthermore, according to any of the aforementioned particular and preferred embodiments wherein the CHA framework structure does not contain $P_2O_5$, and preferably does not contain $Z_2O_5$, in an amount greater than 1 wt.-%, the CHA framework structure, it is further preferred that the CHA framework structure contains $P_2O_5$, and preferably contains $Z_2O_5$, in an amount of 0.5 wt.-% or less, more preferably of 0.1 wt.-% or less, more preferably of 0.05 wt.-% or less, more preferably of 0.01 wt.-% or less, more preferably of 0.005 wt.-% or less, more preferably of 0.001 wt.-% or less, more preferably of 0.0005 wt.-% or less, more preferably of 0.0005 wt.-% or less, more preferably of 0.00005 wt.-% or less, and more preferably of 0.00001 wt.-% or less.

According to particular and preferred embodiments of the present invention wherein the CHA framework structure of the synthetic copper and/or iron containing zeolitic material further comprises $Z_2O_5$ in addition to $SiO_2$ and $X_2O_3$, there is, in general, no particular restriction as to the $YO_2 : X_2O_3$ molar ratios displayed, such that the $YO_2 : X_2O_3$ molar ratio in such inventive materials may have any conceivable value. According to particular embodiments of the present invention wherein the CHA framework structure further comprises $Z_2O_5$ in addition $SiO_2$ and $X_2O_3$, the CHA framework structure then has a $YO_2 : nX_2O_3 : pZ_2O_5$ molar ratio, wherein the value for the ratio $(1+2p) : (n-p)$ may have any conceivable value. Thus, by way of example, the value for the ratio $(1+2p) : (n-p)$ may be 1 or greater than 1, wherein it is preferred that said value is 2 or greater than 2, more preferably 3 or greater than 3, more preferably 5 or greater than 5, and even more preferably 7 or greater than 7. Thus, according to preferred embodiments of the present invention, the value for the ratio $(1+2p) : (n-p)$ of the synthetic copper and/or iron containing zeolitic material having a CHA framework structure further comprising $Z_2O_5$ may range anywhere from 1 to 500, more preferably from 2 to 400, more preferably from 3 to 300, more preferably from 5 to 250, more preferably from 7 to 200, more preferably from 7.5 to 100, more preferably from 8 to 50, more preferably from 8.5 to 30, more preferably from 9 to 20, more preferably from 9.3 to 17, and even more preferably from 9.5 to 15. According to the present invention it is however particularly preferred that in instances wherein the CHA framework structure comprises $Z_2O_5$ in addition to $SiO_2$ and $X_2O_3$, the CHA framework structure has a $SiO_2 : nX_2O_3 : pZ_2O_5$ molar ratio, wherein the value for the ratio $(1+2p) : (n-p)$ is 7 or greater than 7, wherein the value for the ratio $(1+2p) : (n-p)$ ranges from 7.2 to 12, and preferably ranges from 7.4 to 10, more preferably from 7.6 to 9.5, more preferably from 7.8 to 9, more preferably from 7.9 to 8.5, and more preferably from 8 to 8.2.

According to the present invention, there is no particular restriction as to the CHA framework structure displayed by the inventive materials, provided that the copper and/or iron containing zeolitic material having a CHA framework structure displays an X-ray diffraction pattern typical of a CHA framework structure in particular with respect to the reflections and their 2θ degree positions relative to one another. Thus, in principle, provided that the diffraction pattern is typical of a CHA framework structure, there is no particular restriction neither with respect to the actual position of the reflections measured as angle 2θ, nor with respect to the intensities of the individual reflections relative to one another. According to preferred embodiments of the present invention, however, the first reflection in the X-ray diffraction pattern of the copper and/or iron containing zeolitic material having a CHA framework structure, i.e. the reflection having the lowest angle 2θ value is the reflection having highest intensity among all measured reflections, i.e. the reflection measured at the lowest diffraction angle 2θ, has an intensity of 100 %. Furthermore, when using Cu K(alpha 1) radiation, it is further preferred that the diffraction angle 2θ having an intensity of 100 % is comprised in the range of from 5 to 15 °2θ, wherein more preferably, said most intense reflection is comprised in the range of from 8 to 12 °2θ, more preferably of from 9 to 10.5 °2θ, more preferably of from 9.2 to 10 °2θ, more preferably of from 9.5 to 9.7 °2θ, and even more preferably of from 9.55 to 9.65 °2θ. According to particularly preferred embodiments of the present invention, the most intense reflection in the diffraction pattern of the copper and/or iron containing zeolitic material having a CHA framework structure when using Cu K(alpha 1) radiation is comprised in the range of from 9.58 to 9.62 °2θ.

As regards the amount of copper and/or iron contained in the inventive zeolitic material, no particular restrictions apply such that in principle the material may display any conceivable molar ratio (Cu and/or Fe) : $X_2O_3$ of Cu and/or Fe to $X_2O_3$, provided that the zeolitic material contains from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure. Thus, by way of example, the molar ratio (Cu and/or Fe) : $X_2O_3$ of Cu and/or Fe to $X_2O_3$ of the framework structure may range anywhere from 0.05 to 10, wherein preferably the molar ratio ranges from 0.1 to 7, more preferably from 0.5 to 5, more preferably from 1 to 3.5, and more preferably from 1.5 to 3. According to the present invention it is particularly preferred that the molar ratio (Cu and/or Fe) : $X_2O_3$ of Cu and/or Fe to $X_2O_3$ of the framework structure ranges from 1.8 to 2.8.

According to the present invention, the copper and/or iron containing zeolitic material may comprise one or more of any zeolites having a CHA framework structure, provided that said framework structure comprises $SiO_2$ and $X_2O_3$. According to preferred embodiments of the present invention, the copper and/or iron containing zeolitic material comprises one or more zeolites selected from the group consisting of (Ni(deta)$_2$)-UT-6, Chabazite, |Li-Na| [Al-Si-O]-CHA, DAF-5, Na-Chabazite, K-Chabazite, LZ-218, Linde D, Linde R, MeAPSO-47, Phi, SAPO-34, SAPO-47, SSZ-13, SSZ-62, UiO-21, Willhendersonite, ZK-14, ZYT-6, preferably from the group consisting of Chabazite, |Li-Na| [Al-Si-O]-CHA, Na-Chabazite, K-Chabazite, SAPO-34, SAPO-47, SSZ-13, SSZ-62, and combinations of two or more thereof, more preferably from the group consisting of Chabazite, |Li-Na| [Al-Si-O]-CHA, Na-Chabazite, SAPO-34, SSZ-13, and combinations of two or more thereof, more preferably from the group consisting of Na-Chabazite, SAPO-34, SSZ-13, and combinations of two or more thereof, wherein even more preferably the seed crystals having a CHA framework structure comprise Na-Chabazite and/or SAPO-34, and preferably Na-Chabazite.

In addition to relating to a method for the preparation of copper and/or iron containing zeolitic material having a CHA framework structure as such, the present invention further relates to a method for the treatment of $NO_x$ by selective catalytic reduction (SCR) comprising:
(a) providing a catalyst comprising a catalyst according to any of the particular and preferred embodiments of the inventive copper and/or iron containing zeolitic material having a CHA framework structure as defined in the present application; and
(b) contacting a gas stream comprising NOx with the catalyst provided in step (a).

According to the inventive method, it is preferred that the gas stream treated by contacting with the catalyst in (b) comprises one or more reducing agents for selective catalytic reduction of NOx. To this effect, any suitable reducing agent or combination of reducing agents may be employed, provided that they may reduce NOx to nitrogen gas under the catalytic conditions provided by the inventive method. According to the inventive method it is however preferred that urea and/or ammonia is comprised among the reducing agents used, wherein more preferably urea and/or ammonia is employed as the reducing agent in the inventive method, preferably ammonia.

Therefore, it is preferred according inventive method that the gas stream further comprises one or more reducing agents, the one or more reducing agents preferably comprising urea and/or ammonia, preferably ammonia.

As regards the gas comprising NOx, there is no particular restriction according to the present invention as to how said gas stream is provided such that it may stem from any conceivable source. It is, however, preferred according to the present invention that the gas stream comprises a $NO_x$ containing waste gas stream from an internal combustion engine, preferably from an internal combustion engine which operates under lean-burn conditions, and more preferably from a lean-burn gasoline engine or from a diesel engine.

According to the present invention it is alternatively preferred that the gas stream comprises one or more NOx containing waste gases from one or more industrial processes, wherein more preferably the NOx containing waste gas stream comprises one or more waste gas streams obtained in processes for producing adipic acid, nitric acid, hydroxylamine derivatives, caprolactame, glyoxal, methyl-glyoxal, glyoxylic acid or in processes for burning nitrogeneous materials, including mixtures of waste gas streams from two or more of said processes.

Finally, the present invention also relates to the use of the inventive copper and/or iron containing zeolitic material having a CHA framework structure according to any of the particular and preferred embodiments of the present invention, in particular in the field of catalysis and/or in the treatment of exhaust gas, wherein said exhaust gas treatment comprises industrial and automotive exhaust gas treatment. It is, however, preferred according to the present invention that inventive copper and/or iron containing zeolitic material having a CHA framework structure is used as a catalyst for the selective catalytic reduction of NOx, and preferably in the treatment of NOx containing exhaust gas by SCR, wherein more preferably the zeolitic material is used in the treatment of industrial or automotive exhaust gas. According to the present invention it is particularly preferred that the inventive copper and/or iron containing zeolitic material having a CHA framework structure according to any of the particular and preferred embodiments defined in the present application is used in the treatment of automotive exhaust gas.

The present invention is further characterized by the following and particular preferred embodiments, including the combination and embodiments indicated by the respective dependencies:

1. An organotemplate-free synthetic process for the production of a copper and/or iron containing zeolitic material having a CHA framework structure comprising $YO_2$, $X_2O_3$, and optionally comprising $Z_2O_5$, wherein said process comprises:

(1) preparing a mixture comprising one or more sources for $YO_2$, one or more sources for $X_2O_3$, and seed crystals having a CHA framework structure, wherein the CHA framework structure of the seed crystals comprises $YO_2$, $X_2O_3$, and optionally comprises $Z_2O_5$;
(2) crystallizing the mixture prepared in (1) for obtaining a zeolitic material having a CHA framework structure; and
(3) subjecting the zeolitic material obtained in (2) to one or more ion exchange procedures for obtaining a zeolitic material ion exchanged with Cu and/or Fe, preferably with Cu;
wherein Y is a tetravalent element, X is a trivalent element, and Z is a pentavalent element,
wherein the ion exchanged zeolitic material obtained in (3) contains from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $YO_2$ contained in the zeolitic material having a CHA framework structure.

2. The process of embodiment 1, wherein the metal ion exchanged zeolitic material obtained in (3) contains from 4.5 to 10 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $YO_2$ contained in the zeolitic material having a CHA framework structure, preferably from 5.2 to 9.5 wt.-%, more preferably from 5.5 to 9 wt.-%, more preferably from 6 to 8.5 wt.-%, more preferably from 6.5 to 8 wt.-%, and more preferably from 7 to 7.5 wt.-%.

3. The process of embodiment 1 or 2, wherein optionally one or more sources for $Z_2O_5$ are further provided in (1), and
wherein if the CHA framework structure of the seed crystals does not contain $Z_2O_5$, the seed crystals then have a $YO_2 : X_2O_3$ molar ratio of 5 or greater than 5.

4. The process of any of embodiments 1 to 3, wherein (3) comprises one or more of the following
(3a) subjecting the zeolitic material obtained in (2) to one or more ion exchange procedures with $H^+$ and/or $NH_4^+$, preferably with $NH_4^+$;
(3b) optionally calcining the zeolitic material obtained in (3a); and
(3c) subjecting the zeolitic material obtained in (3b) to one or more ion exchange procedures with Cu and/or Fe.

5. The process of embodiment 4, wherein calcining of the zeolitic material in (3b) is effected by heating of the zeolitic material to a temperature ranging from 200 to 850°C, preferably from 250 to 750°C, more preferably from 300 to 700°C, more preferably from 350 to 650 °C, more preferably from 400 to 600°C, more preferably from 450 to 550°C, and more preferably from 475 to 525°C.

6. The process of embodiment 4 or 5, wherein calcining of the zeolitic material in (3b) is effected by heating of the zeolitic material for a duration ranging from 0.5 to 36 h, preferably from 1 to 24 h, more preferably from 2 to 20 h, more preferably from 3 to 16 h, more preferably from 3.5 to 12 h, more preferably from 4 to 10 h, more preferably from 4.5 to 8 h, more preferably from 5 to 7 h, and more preferably from 5.5 to 6.5 h.

7. The process of any of embodiments 1 to 6, wherein if the CHA framework structure of the seed crystals further comprises $Z_2O_5$ in addition to $YO_2$ and $X_2O_3$, the seed crystals then have a $YO_2 : nX_2O_3 : pZ_2O_5$ molar ratio, wherein the value for the ratio $(1+2p) : (n-p)$ is 5 or greater than 5, wherein the value for the ratio $(1+2p) : (n-p)$ preferably ranges from 5 to 200, more preferably from 6 to 150, more preferably from 8 to 100, more preferably from 12 to 70, more preferably from 20 to 50, more preferably from 25 to 40, more preferably from 28 to 35, and even more preferably from 29 to 33.

8. The process of any of embodiments 1 to 7, wherein the $YO_2 : X_2O_3$ molar ratio of the mixture prepared in (1) ranges from 1 to 150, preferably from 2 to 100, more preferably from 5 to 70, more preferably from 10 to 50, more preferably from 13 to 30, more preferably from 16 to 25, and even more preferably from 18 to 22.

9. The process of any of embodiments 1 to 8, wherein the mixture prepared in (1) comprises one or more alkali metals M, wherein the one or more alkali metals M are preferably selected from the group consisting of Li, Na, K, and combinations of two or more thereof, wherein more preferably the one or more alkali metals are Li and/or Na, and preferably Na.

10. The process of embodiment 9, wherein the $M_2O : YO_2$ molar ratio in the mixture according to (1) ranges from 0.01 to 5, preferably from 0.05 to 2, more preferably from 0.1 to 1.5, more preferably from 0.15 to 1, more preferably from 0.2 to 0.5, and even more preferably from 0.25 to 0.35.

11. The process of embodiment 9 or 10, wherein the $YO_2 : X_2O_3 : M_2O$ molar ratios in the mixture prepared in (1) range from (5 - 70) : 1 : (0.5 - 20), preferably from (10 - 50) : 1 : (1 - 15), more preferably from (13 - 30) : 1 : (2 - 10), more preferably from (16 - 25) : 1 : (4 - 8), and even more preferably from (18 - 22) : 1 : (5 - 7).

12. The process of any of embodiments 1 to 11, wherein the mixture prepared in (1) contains no K and/or no Sr, preferably no K.

13. The process of any of embodiments 1 to 12, wherein the seed crystals having a CHA framework structure display an X-ray diffraction pattern wherein the most intense reflec-

tion having an intensity of 100% has a diffraction angle 2θ in the range of from 5 to 15, preferably of from 8 to 12, more preferably of from 9 to 10.5, more preferably of from 9.2 to 10, more preferably of from 9.5 to 9.7, more preferably of from 9.55 to 9.65, and even more preferably of from 9.58 to 9.62,
wherein the diffraction angle 2θ is obtained using Cu K(alpha 1) radiation.

14. The process of any of embodiments 1 to 13, wherein Y comprised in the seed crystals and/or, preferably and,
wherein Y provided in (1) in the one or more sources for $YO_2$ are, independently from one another, selected from the group consisting of Si, Sn, Ti, Zr, Ge, and mixtures of two or more thereof, Y preferably being Si.

15. The process of any of embodiments 1 to 14, wherein the one or more sources for $YO_2$ comprises one or more silicates, preferably one or more alkali metal silicates, wherein the alkali metal is preferably selected from the group consisting of Li, Na, K, and combinations of two or more thereof, wherein more preferably the alkali metal is Na and/or K, and wherein even more preferably the alkali metal is Na.

16. The process of embodiment 15, wherein the mixture prepared in (1) preferably further comprises silica in addition to the one or more silicates, preferably one or more silica hydrosols and/or one or more colloidal silicas, and even more preferably one or more colloidal silicas in addition to the one or more silicates.

17. The process of embodiment 15 or 16, wherein the mixture prepared in (1) comprises water glass, preferably sodium and/or potassium silicate, more preferably sodium silicate.

18. The process of any of embodiments 1 to 17, wherein X comprised in the seed crystals and/or, preferably and,
wherein X provided in (1) in the one or more sources for $X_2O_3$ are, independently from one another, selected from the group consisting of Al, B, In, Ga, and mixtures of two or more thereof, X preferably being Al and/or B, and more preferably being Al.

19. The process of any of embodiments 1 to 18, wherein the one or more sources for $X_2O_3$ comprises one or more aluminate salts, preferably on or more alkali metal aluminates, wherein the alkali metal is preferably selected from the group consisting of Li, Na, K, and combinations of two or more thereof, wherein more preferably the alkali metal is Na and/or K, and wherein even more preferably the alkali metal is Na.

20. The process of any of embodiments 1 to 19, wherein Z optionally comprised in the seed crystals
    and/or, preferably and,
    wherein Z in the one or more sources for $Z_2O_5$ optionally further provided in (1) are, independently from one another, selected from the group consisting of P, As, Sb, Bi, V, Nb, Ta, and combinations of two or more thereof, preferably from the group consisting of P, As, V, and combinations of two or more thereof, wherein more preferably Z comprises P or As, preferably P, and wherein even more preferably Z is P.

21. The process of any of embodiments 1 to 20, wherein the one or more sources for $Z_2O_5$ comprises one or more phosphates and/or one or more oxides and/or one or more acids of phosphorous, preferably one or more acids of phosphorous, more preferably phosphoric acid, and wherein even more preferably the source for $Z_2O_5$ is phosphoric acid.

22. The process of any of embodiments 1 to 21, wherein the seed crystals having a CHA framework structure comprise one or more zeolites selected from the group consisting of $(Ni(deta)_2)$-UT-6, Chabazite, |Li-Na| [Al-Si-O]-CHA, DAF-5, Na-Chabazite, K-Chabazite, LZ-218, Linde D, Linde R, MeAPSO-47, Phi, SAPO-34, SAPO-47, SSZ-13, SSZ-62, UiO-21, Willhendersonite, ZK-14, ZYT-6, and combinations of two or more thereof,
    preferably from the group consisting of Chabazite, |Li-Na| [Al-Si-O]-CHA, Na-Chabazite, K-Chabazite, SAPO-34, SAPO-47, SSZ-13, SSZ-62, and combinations of two or more thereof,
    more preferably from the group consisting of Chabazite, |Li-Na| [Al-Si-O]-CHA, Na-Chabazite, SAPO-34, SSZ-13, and combinations of two or more thereof,
    more preferably from the group consisting of Na-Chabazite, SAPO-34, SSZ-13, and combinations of two or more thereof,
    wherein even more preferably the seed crystals having a CHA framework structure comprise Na-Chabazite and/or SAPO-34, and preferably Na-Chabazite.

23. The process of any of embodiments 1 to 22, wherein the amount of seed crystals in the mixture prepared in (1) ranges from 0.1 to 35 wt.-% based on 100 wt.-% of $YO_2$ in the one or more sources for $YO_2$, preferably from 0.5 to 28 wt.-%, more preferably from 1 to 22 wt.-%, more preferably from 5 to 18 wt.-%, more preferably from 10 to 15 wt.-%, and even more preferably from 12 to 14 wt.-%.

24. The process of any of embodiments 1 to 23, wherein the mixture prepared in (1) further comprises one or more solvents, wherein said one or more solvents preferably comprises water, more preferably distilled water.

25. The process of embodiment 24, wherein the mixture prepared in (1) further comprises water as a solvent, and wherein the $H_2O : YO_2$ molar ratio of the mixture prepared in (1) ranges from 1 to 25, preferably from 2 to 20, more preferably from 4 to 18, more preferably from 6 to 16, more preferably from 8 to 14, and even more preferably from 10 to 12.

26. The process of embodiment 24 or 25, wherein the crystallization in (2) is conducted under solvothermal conditions, preferably under hydrothermal conditions.

27. The process of any of embodiments 1 to 25, wherein the crystallization in (2) involves heating of the mixture, preferably at a temperature ranging from 80 to 200°C, more preferably from 90 to 180°C, more preferably from 100 to 160°C, more preferably from 110 to 140°C, and even more preferably from 115 to 130°C.

28. The process of any of embodiments 1 to 27, wherein the crystallization in (2) involves heating of the mixture for a period ranging from 2 to 36 h, more preferably from 6 to 24 h, more preferably from 12 to 20 h, and even more preferably from 14 to 18 h.

29. The process of any of embodiments 1 to 28, (2) further comprising one or more of the following
(2a) isolating the zeolitic material having a CHA framework structure, preferably by filtration,
and/or
(2b) washing the zeolitic material having a CHA framework structure,
and/or
(2c) drying the zeolitic material having a CHA framework structure,
wherein the steps (2a) and/or (2b) and/or (2c) can be conducted in any order, and wherein one or more of said steps is preferably repeated one or more times.

30. The process of any of embodiments 4 to 29, (3a) further comprising one or more of the following
(3a.i) isolating the zeolitic material ion exchanged with $H^+$ and/or $NH_4^+$, preferably by filtration,
and/or
(3a.ii) washing the zeolitic material ion exchanged with $H^+$ and/or $NH_4^+$,,
and/or
(3a.iii) drying the zeolitic material ion exchanged with $H^+$ and/or $NH_4^+$,
wherein the steps (3a.i) and/or (3a.ii) and/or (3a.iii) can be conducted in any order, and wherein one or more of said steps is preferably repeated one or more times.

31. The process of any of embodiments 4 to 30, (3c) further comprising one or more of the following
(3c.i) isolating the zeolitic material ion exchanged with Cu and/or Fe, preferably by filtration,
and/or
(3c.ii) washing the zeolitic material ion exchanged with Cu and/or Fe,
and/or
(3c.iii) drying the zeolitic material ion exchanged with Cu and/or Fe,
wherein the steps (3c.i) and/or (3c.ii) and/or (3c.iii) can be conducted in any order, and wherein one or more of said steps is preferably repeated one or more times.

32. The process of any of embodiments 1 to 31, wherein in (3) at least one ionic non-framework element contained in the zeolitic material having a CHA framework is ion exchanged against Cu and/or Fe, and preferably against Cu,
wherein said at least one ionic non-framework element is preferably one or more alkali metals, more preferably Na and/or K, and more preferably Na.

33. The process of any of embodiments 1 to 32, wherein prior to the one or more ion exchange procedures, the organotemplate-free synthetic process does not comprise a calcination procedure.

34. The process of any of embodiments 1 to 33, wherein the seed crystals comprise a zeolitic material having a CHA framework structure as obtained in (2) according to the process of any of embodiments 1 to 33.

35. A synthetic copper and/or iron containing zeolitic material having a CHA framework structure obtainable and/or obtained according to the process of any of embodiments 1 to 34.

36. A synthetic copper and/or iron containing zeolitic material having a CHA framework structure, optionally obtainable and/or obtained according to the process of any of embodiments 1 to 34, wherein the CHA framework structure comprises $SiO_2$, $X_2O_3$, and optionally comprises $Z_2O_5$, wherein X is a trivalent element, and Z is a pentavalent element,
wherein the zeolitic material contains from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure, and
wherein the $^{29}Si$ MAS NMR of the zeolitic material comprises:
a first peak (P1) in the range of from −96 to −98.8 ppm, preferably of from −96.5 to −98.5 ppm, more preferably of from −96.8 to −98. 2 ppm, more preferably of from −97 to −98 ppm, more preferably of from −97.3 to −97.9 ppm, and even more preferably of from −97.5 to −97.7 ppm;

a second peak (P2) in the range of from −102 to −104.5 ppm, preferably of from −102.5 to −104 ppm, more preferably of from −102.8 to −103.7 ppm, more preferably of from −103 to −103.5 ppm, and even more preferably of from −103.2 to −103.4 ppm; and

a third peak (P3) in the range of from −107.5 to −111 ppm, preferably of from −108 to −110.5 ppm, more preferably of from −108.5 to −110 ppm, more preferably of from −108.8 to −109.5 ppm, more preferably of from −109 to −109.4 ppm, and even more preferably of from −109.1 to −109.3 ppm;

wherein the integration of the first, second, and third peaks in the $^{29}$Si MAS NMR of the zeolitic material offers a ratio of the integration values P1 : P2 : P3 ranging from (0.35 – 0.7) : 1 : (0.1 – 1.6), preferably from (0.4 – 0.65) : 1 : (0.15 – 1.3), more preferably from (0.42 – 0.62) : 1 : (0.2 – 1), more preferably from (0.45 – 0.6) : 1 : (0.3 – 0.7), more preferably from (0.48 – 0.58) : 1 : (0.35 – 0.5), more preferably from (0.5 – 0.56) : 1 : (0.4 – 0.45), and even more preferably from (0.52 – 0.54) : 1 : (0.42 – 0.44).

37. The zeolitic material of embodiment 36, wherein the zeolitic material contains from 4.5 to 10 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure, preferably from 5.2 to 9.5 wt.-%, more preferably from 5.5 to 9 wt.-%, more preferably from 6 to 8.5 wt.-%, more preferably from 6.5 to 8 wt.-%, and more preferably from 7 to 7.5 wt.-%.

38. The zeolitic material of embodiment 36 or 37, wherein X is selected from the group consisting of Al, B, In, Ga, and mixtures of two or more thereof, X preferably being Al and/or B, and more preferably being Al.

39. The zeolitic material of any of embodiments 36 to 38, wherein the CHA framework structure does not contain $Z_2O_5$ in an amount greater than 1 wt.-% based on 100 wt.-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure, and wherein the CHA framework structure has an $SiO_2 : X_2O_3$ molar ratio of 7 or greater than 7, and preferably ranging from 7.2 to 12, more preferably from 7.4 to 10, more preferably from 7.6 to 9.5, more preferably from 7.8 to 9, more preferably from 7.9 to 8.5, and more preferably from 8 to 8.2.

40. The zeolitic material of any of embodiments 36 to 39, wherein the CHA framework structure comprises $Z_2O_5$ in addition to $SiO_2$ and $X_2O_3$, and the CHA framework structure has a $YO_2 : nX_2O_3 : pZ_2O_5$ molar ratio, wherein the value for the ratio $(1+2p) : (n-p)$ is 7 or greater than 7, wherein the value for the ratio $(1+2p) : (n-p)$ preferably ranges from 7.2 to 12, more preferably from 7.4 to 10, more preferably from 7.6 to 9.5, more preferably from 7.8 to 9, more preferably from 7.9 to 8.5, and more preferably from 8 to 8.2.

41. The zeolitic material of any of embodiments 36 to 40, wherein Z is selected from the group consisting of P, As, Sb, Bi, V, Nb, Ta, and combinations of two or more thereof, preferably from the group consisting of P, As, V, and combinations of two or more thereof, wherein more preferably Z comprises P or As, preferably P, and wherein even more preferably Z is P.

42. The zeolitic material of any of embodiments 36 to 41, wherein the CHA framework structure displays an X-ray diffraction pattern wherein the most intense reflection having an intensity of 100% has a diffraction angle 2θ in the range of from 5 to 15, preferably of from 8 to 12, more preferably of from 9 to 10.5, more preferably of from 9.2 to 10, more preferably of from 9.5 to 9.7, more preferably of from 9.55 to 9.65, and even more preferably of from 9.58 to 9.62, wherein the diffraction angle 2θ is obtained using Cu K(alpha 1) radiation.

43. The zeolitic material of any of embodiments 36 to 42, wherein the CHA framework comprises at least a portion of the Cu and/or Fe as extra-framework ions.

44. The zeolitic material of any of embodiments 36 to 43, wherein the Cu and/or Fe has at least in part been introduced into the zeolitic material via ion exchange.

45. The zeolitic material of embodiment 44, wherein the molar ratio (Cu and/or Fe) : $X_2O_3$ of Cu and/or Fe to $X_2O_3$ of the framework structure ranges from 0.05 to 10, preferably from 0.1 to 7, more preferably from 0.5 to 5, more preferably from 1 to 3.5, more preferably from 1.5 to 3, and even more preferably from 1.8 to 2.8.

46. The zeolitic material of any of embodiments 36 to 45, wherein the zeolitic material comprises one or more zeolites selected from the group consisting of (Ni(deta)$_2$)-UT-6, Chabazite, |Li-Na| [Al-Si-O]-CHA, DAF-5, Na-Chabazite, K-Chabazite, LZ-218, Linde D, Linde R, MeAPSO-47, Phi, SAPO-34, SAPO-47, SSZ-13, SSZ-62, UiO-21, Willhendersonite, ZK-14, ZYT-6, and combinations of two or more thereof,
preferably from the group consisting of Chabazite, |Li-Na| [Al-Si-O]-CHA, Na-Chabazite, K-Chabazite, SAPO-34, SAPO-47, SSZ-13, SSZ-62, and combinations of two or more thereof,
more preferably from the group consisting of Chabazite, |Li-Na| [Al-Si-O]-CHA, Na-Chabazite, SAPO-34, SSZ-13, and combinations of two or more thereof,
more preferably from the group consisting of Na-Chabazite, SAPO-34, SSZ-13, and combinations of two or more thereof,
wherein even more preferably the seed crystals having a CHA framework structure comprise Na-Chabazite and/or SAPO-34, and preferably Na-Chabazite.

47. Method for the treatment of $NO_x$ by selective catalytic reduction (SCR) comprising:
    (a) providing a catalyst comprising a zeolitic material according to any of embodiments 35 to 46; and
    (b) contacting a gas stream comprising $NO_x$ with the catalyst provided in (a).

48. The method of embodiment 47, wherein the gas stream further comprises one or more reducing agents, the one or more reducing agents preferably comprising urea and/or ammonia, preferably ammonia.

49. The method of embodiment 47 or 48, wherein the gas stream comprises one or more $NO_x$ containing waste gases, preferably one or more $NO_x$ containing waste gases from one or more industrial processes, wherein more preferably the $NO_x$ containing waste gas stream comprises one or more waste gas streams obtained in processes for producing adipic acid, nitric acid, hydroxylamine derivatives, caprolactame, glyoxal, methyl-glyoxal, glyoxylic acid or in processes for burning nitrogeneous materials, including mixtures of waste gas streams from two or more of said processes.

50. The method of any of embodiments 47 to 49, wherein the gas stream comprises a $NO_x$ containing waste gas stream from an internal combustion engine, preferably from an internal combustion engine which operates under lean-burn conditions, and more preferably from a lean-burn gasoline engine or from a diesel engine.

51. Use of a synthetic organotemplate-free zeolitic material having a CHA framework structure according to any of embodiments 35 to 46 as a molecular sieve, as an adsorbent, for ion exchange, as a catalyst and/or as a catalyst support, preferably as a catalyst, more preferably as a catalyst in the selective catalytic reduction (SCR) of $NO_x$, more preferably as a catalyst in the treatment of exhaust gas by SCR, more preferably in the treatment of $NO_x$ containing automotive or industrial exhaust gas by SCR, preferably of automotive exhaust gas.

## DESCRIPTION OF THE FIGURES

The X-ray diffraction (XRD) patterns shown in the Figures were respectively measured using Cu K alpha-1 radiation. In the respective diffractograms, the diffraction angle 2 theta in ° is shown along the abscissa and the intensities are plotted along the ordinate.

Figure 1a shows the X-ray diffraction pattern of the zeolitic material obtained from Reference Example 1. For comparative purposes, the line pattern of the CHA type framework structure is indicated in the diffractogram.

Figures 1b and 2a show the $^{29}$Si MAS NMR spectrum obtained for the zeolitic materials of Reference Examples 1 and 2, respectively.

Figures 1c and 2b show the $^{27}$Si MAS NMR spectrum obtained for the zeolitic materials of Reference Examples 1 and 2, respectively.

Figure 3 shows the results from SCR catalytic testing obtained for the zeolitic materials of Example 1 and Comparative Example 1 at different copper loadings. In the figure, the copper loading in wt.-% (of Cu calculated as the element) of the zeolitic material for the different samples is shown along the abscissa and the NOx conversion rate in % is plotted along the ordinate. The results for the fresh samples from Example 1 tested at 200°C are indicated by “◆”, the fresh samples from Comparative Example 1 tested at 200°C are indicated by “●”, the fresh samples from Example 1 tested at 575°C are indicated by “▲”, and the fresh samples from Comparative Example 1 tested at 575°C are indicated by “X”.

Figure 4 shows the results from SCR catalytic testing obtained for the zeolitic materials of Example 1 and Comparative Example 1 at different copper loadings after aging for 5 h at 750°C. In the figure, the copper loading in wt.-% (of Cu calculated as the element) of the zeolitic material for the different samples is shown along the abscissa and the NOx conversion rate in % is plotted along the ordinate. The results for the aged samples from Example 1 tested at 200°C are indicated by “◆”, the aged samples from Comparative Example 1 tested at 200°C are indicated by “●”, the aged samples from Example 1 tested at 575°C are indicated by “▲”, and the aged samples from Comparative Example 1 tested at 575°C are indicated by “X”.

## EXAMPLES

In the following examples, $^{29}$Si and $^{27}$Al solid-state NMR experiments were conducted on a Avance 300 spectrometer equipped with a wide-bore magnet (Bruker Biospin, Germany).

For $^{29}$Si solid-state NMR, samples were packed in 7 mm rotors and measured under 5 kHz Magic Angle Spinning at room temperature. $^{29}$Si spectra were recorded using direct polarization with a 90° pulse, 25 ms signal acquisition under high-power proton heteronuclear decoupling, and a recycle delay of 120 s. Spectra were referenced with the polymer Q8M8 as an external secondary standard, setting the resonance of the trimethylsilyl M group to 12.5 ppm. The spectra were processed with Bruker Topspin (Bruker Biospin, Germany).

For $^{27}Al$ solid-state NMR, samples were packed in 4 mm rotors and measured under 10 kHz Magic Angle Spinning at room temperature. $^{27}Al$ spectra were recorded using direct polarization with a 15° pulse, 20 ms signal acquisition, and a recycle 0.25 s. Spectra were referenced with 1 mol/L $AlCl_3$•$6H_2O$ solution as zero reference. The spectra were processed with Bruker Topspin (Bruker Biospin, Germany).

**Reference Example 1:** Organotemplate-free synthesis of chabazite

In a reaction vessel 6.75 kg $NaAlO_2$ were dissolved in 46.12 kg $H_2O$ (DI). Under stirring, 6.37 kg Chabazite seed crystals are then dispersed followed by the slow addition of 119.99 kg waterglass (26 wt-% $SiO_2$, 8 wt-% $Na_2O$, 66 wt-% $H_2O$). Finally, 28.12 kg LUDOX AS 40 ($SiO_2$ wt-% in H2O) is given in the stirred reaction mixture. The resulting reaction gel accordingly displays an $SiO_2$ : $Al_2O_3$ : $Na_2O$ : $H_2O$ molar ratio of 36 : 1.8 : 10.3 : 400. The reaction mixture is then transferred into a static autoclave and is heated for 60 h to 120°C. Afterwards the dispersion is cooled down and the solid is separated from the supernatant by filtration and subsequent washing with $H_2O$ (DI) until a conductivity of 200 µS is reached. In order to fully remove residual $H_2O$, the sample was dried for 16 h at 120°C in a static oven under air. 27 kg of a white powder were obtained.

Elemental analysis of the product afforded 75.3 wt-% $SiO_2$, 15.8 wt-% $Al_2O_3$ and 8.8 wt-% $Na_2O$.

Nitrogen sorption of the product at 77K afforded a specific surface area of 199 $m^2$/g.

As may be taken from the X-ray diffraction of the obtained product displayed in Figure 1a, the product reveals a zeolitic material having CHA framework structure.

In Figure 1b, the $^{29}Si$ MAS NMR of the crystalline product obtained from the synthetic procedure is shown. As may be taken from the NMR spectrum, three major peaks are observed at −97.6 ppm (P1), −103.3 ppm (P2), and −109.2 ppm (P3), respectively, wherein the relative instensity of the peaks afford a P1 : P2 : P3 ratio of 0.268 : 0.515 : 0.216. In particular, the signal at -109 ppm of the $^{29}Si$ MAS NMR corresponds to Q4 structures, wherein the respective signals at -103 and -98 ppm are attributed to the Q3 or to Q4 structures.

In Figure 1c, the $^{27}Al$ MAS NMR spectrum of the crystalline product is displayed, revealing a peak at 58.4 ppm corresponding to tetrahedrally coordinated Al sites of the chabazite material obtained from organotemplate free synthesis. There is no measurable intensity in the -5 to -15 ppm range which could be attributed to octahedrally coordinated aluminum contained in the sample.

**Reference Example 2:** Synthesis of chabazite using an organotemplate

174.6 g $H_2O$ were stirred together with 478.8 g of a 20wt-% Adamantyltrimethylammoniumhydroxide (organic template) solution in $H_2O$. 102.7 g NaOH (25 wt-% in $H_2O$) were added slowly under stirring. After 10 minutes 80.4 g Aluminiumtriisopropylate were dissolved in the reaction mixture followed by the addition of 963.4g LUDOX AS 40 after 60 minutes. The resulting reaction gel accordingly displays an $SiO_2$ : $Al_2O_3$ : $Na_2O$ : $H_2O$ : organotemplate molar ratio of 36 : 2.2 : 3.6 : 377 : 2.6. Finally the reaction mixture is stirred for 30 min before it is placed into an autoclave, which is heated to 170°C for 20 hours. After the autoclave was cooled to room temperature, the resulting dispersion was adjusted by means of a 10wt-% $HNO_3$ solution in $H_2O$ to a pH-value of 7. Afterwards to the resulting solid was filtered and washed with $H_2O$ until a conductivity of below 200 µS is reached. Afterwards the solid was first dried at 120°C for 10h and then calcined under air at 600°C for 6h. From this approach 391g white powder were obtained having an elemental composition of 80.3 wt-% $SiO_2$, 4.4 wt-% $Al_2O_3$ and 0.7 wt-% $Na_2O$ and 13.8 wt-% C. Nitrogen sorption at 77K indicated a specific surface area of 70 $m^2/g$.

Elemental analysis of the product afforded 80.3 wt-% $SiO_2$, 4.4 wt-% $Al_2O_3$ and 0.7 wt-% $Na_2O$ and 13.8 wt-% C.

Nitrogen sorption of the product at 77K afforded a specific surface area of 70 $m^2/g$.

In Figure 2a, the $^{29}Si$ MAS NMR of the crystalline product obtained from the synthetic procedure is shown. As may be taken from the NMR spectrum, three major peaks are observed at −98.8 ppm (P1), −104.1 ppm (P2), and −109.9 ppm (P3), respectively, wherein the relative instensity of the peaks afford a P1 : P2 : P3 ratio of 0.137 : 0.412 : 0.442. In particular, the signal at -110 ppm of the $^{29}Si$ MAS NMR corresponds to Q4 structures, wherein the respective signals at -104 and -99 ppm are attributed to the Q3 or to Q4 structures.

In Figure 2b, the $^{27}Al$ MAS NMR spectrum of the crystalline product is displayed, revealing a peak at 57.5 ppm corresponding to tetrahedrally coordinated Al and a peak at -2.2 ppm corresponding to octahedrally coordinated Al sites of the chabazite material.

**Example 1:** Loading of Reference Example 1 with copper

27.0 kg of the zeolite powder obtained from Reference Example 1 were dispersed in a solution of 26.6 kg $NH_4NO_3$ in 265.8 kg $H_2O$ (DI). Under stirring, the mixture was heated for 2 hours to 80°C. Then, the solid was separated from the aqueous phase by filtration and subsequent washing with $H_2O$ until no nitrate can be detected in the washing water. The obtained white solid powder was dried at 120°C for 16 h under air. Afterwards the obtained powder was treated at 500°C for 6 hours under air in order to transfer the zeolite in the H-Form.

Nitrogen sorption at 77K afforded a specific surface area of 571 $m^2/g$ for the calcined material.

Respective samples of the H-form as obtained after calcination were then loaded via incipient wetness impregnation with various $Cu(NO_3)_2$ solutions for obtaining copper loadings ranging from 1.0 wt-% to 7.5 wt-% Cu calculated as the element. The impregnated powders were placed in sealed beakers for 20 hours at 50°C. Afterwards, the materials were calcined at 5h for 450°C under air.

**Comparative Example 1:** Loading of Reference Example 2 with copper

The procedure of Example 1 was repeated with Reference Example 2 for affording a copper loaded comparative example displaying a copper loading ranging from 1.0 wt-% to 7.5 wt-% Cu calculated as the element.

Nitrogen sorption performed at 77K on the H-form of the zeolitic material after ion exchange with ammonium and subsequent calcination afforded a specific surface area of 623 $m^2/g$.

**Example 2:** SCR Testing

The copper loaded materials obtained from Example 1 and Comparative Example 1 were respectively dispersed in $H_2O$ in which 5 wt-% of $Zr(acetate)_2$ based on the amount of zeolite is added. The slurrys were then dried under stirring and calcined afterwards at 600°C for 1 hour under air. The obtained solids were crushed and sieved to a particle size of 250 – 500 µm for catalytic testing.

The respective samples were subsequently tested under selective catalytic reduction conditions relative to their NOx conversion capacity. To this effect the samples were contacted at 200°C and 575°C with a gas stream containing 500 ppm nitrogen oxide, 500 ppm ammonia, 5 volume percent water, 10 volume percent oxygen (as air) and balance nitrogen at a weight hourly space velocity (WHSV) of 80,000 $h^{-1}$. The samples were then aged at 750°C for 5 hours in an atmosphere containing 10 volume percent of water, and then tested anew. The results of said testing are displayed in Figures 3 and 4.

As may be taken from the results from comparative testing displayed in Figure 3, at conventional copper loadings up to 3.5 wt.-% copper, the conventional catalysts containing chabazite obtained from a synthesis route using an organotemplate afford an optimum in conversion rate. Increasing the loading in copper leads to a steady decline in $NO_x$ conversion levels at both low

(200°C) and high (575°C) temperatures. As may be taken from the results displayed in Figure 4, same applies relative to the aged catalysts, wherein the decline in $NO_x$ conversion activity with increasing copper loadings is accentuated.

With respect to the catalyst samples containing chabazite obtained from organotemplate-free synthesis according to Reference Example 1, it may be observed that the fresh catalysts display a higher conversion rate at high temperatures and a slightly lower conversion rate at low temperatures up to the optimum loading of 3.5 wt.-% found for the conventional catalysts containing chabazite obtained from organotemplate-mediated synthesis. After aging, however, a high degradation in the conversion rate at low temperatures is observed for those copper loading levels for the samples containing chabazite from organotemplate-free synthesis.

Accordingly, an advantage for using copper chabazite obtained from organotemplate-free synthesis was not readily apparent at conventional copper loadings. Furthermore, in view of the generally lower $SiO_2$ : $Al_2O_3$ molar ratio in chabazite obtained from organotemplate-free synthesis, it was expected that such catalysts would behave only poorly after aging due to the high levels of alumina in the framework structure, and as observed in Figure 4 for the aged catalyst when tested at lower temperatures. It has, however, quite unexpectedly been found that the catalysts containing chabazite from organotemplate-free synthesis display an improved catalytic activity in the conversion of $NO_x$ when applying high levels of copper loading. Even more surprisingly, it has been found that high loadings of copper apparently stabilize chabazite obtained from organotemplate-free synthesis since with increasing copper loadings the aged samples actually show improved $NO_x$ conversion levels, in particular at low temperatures (see Figure 4). In fact, it has unexpectedly been found that by increasing the copper loading in chabazite obtained from organotemplate-free synthesis, the $NO_x$ conversion may even clearly outperform conventional copper chabazite materials obtained from organotemplate-mediated synthetic procedures, as may for example be observed in Figures 3 and 4 for samples containing chabazite obtained from organotemplate-free synthesis at a copper loading of about 6 wt.-% compared to the optimum levels of conversion obtained with conventional copper chabazite at copper loadings of around 3.5 wt.-%.

Therefore, it has quite surprisingly been found that a clearly improved catalyst for the conversion of $NO_x$ by selective catalytic reduction may be provided when employing chabazite from organotemplate-free synthesis loaded with high levels of copper. Even more unexpectedly, it has been found that such improved levels of $NO_x$ conversion may be achieved even after aging of the catalyst, despite the high alumina content of the framework structure.

## Claims

1. An organotemplate-free synthetic process for the production of a copper and/or iron containing zeolitic material having a CHA framework structure comprising $YO_2$, $X_2O_3$, and optionally comprising $Z_2O_5$, wherein said process comprises:
(1) preparing a mixture comprising one or more sources for $YO_2$, one or more sources for $X_2O_3$, and seed crystals having a CHA framework structure, wherein the CHA framework structure of the seed crystals comprises $YO_2$, $X_2O_3$, and optionally comprises $Z_2O_5$;
(2) crystallizing the mixture prepared in (1) for obtaining a zeolitic material having a CHA framework structure; and
(3) subjecting the zeolitic material obtained in (2) to one or more ion exchange procedures for obtaining a zeolitic material ion exchanged with Cu and/or Fe, preferably with Cu;
wherein Y is a tetravalent element, X is a trivalent element, and Z is a pentavalent element,
wherein the ion exchanged zeolitic material obtained in (3) contains from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $YO_2$ contained in the zeolitic material having a CHA framework structure.

2. The process of claim 1, wherein the $YO_2$ : $X_2O_3$ molar ratio of the mixture prepared in (1) ranges from 1 to 150.

3. The process of claim 1 or 2, wherein Y comprised in the seed crystals
and/or, preferably and,
wherein Y provided in (1) in the one or more sources for $YO_2$ are, independently from one another, selected from the group consisting of Si, Sn, Ti, Zr, Ge, and mixtures of two or more thereof.

4. The process of any of claims 1 to 3, wherein X comprised in the seed crystals
and/or, preferably and,
wherein X provided in (1) in the one or more sources for $X_2O_3$ are, independently from one another, selected from the group consisting of Al, B, In, Ga, and mixtures of two or more thereof.

5. The process of any of claims 1 to 4, wherein Z optionally comprised in the seed crystals
and/or, preferably and,
wherein Z in the one or more sources for $Z_2O_5$ optionally further provided in (1) are, independently from one another, selected from the group consisting of P, As, Sb, Bi, V, Nb, Ta, and combinations of two or more thereof.

6. The process of any of claims 1 to 5, wherein the mixture prepared in (1) further comprises one or more solvents.

7. The process of claim 6, wherein the mixture prepared in (1) further comprises water as a solvent, and wherein the $H_2O : YO_2$ molar ratio of the mixture prepared in (1) ranges from 1 to 25.

8. The process of claim 6 or 7, wherein the crystallization in (2) is conducted under solvothermal conditions.

9. A synthetic copper and/or iron containing zeolitic material having a CHA framework structure obtainable and/or obtained according to the process of any of claims 1 to 8.

10. A synthetic copper and/or iron containing zeolitic material having a CHA framework structure, optionally obtainable and/or obtained according to the process of any of claims 1 to 8, wherein the CHA framework structure comprises $SiO_2$, $X_2O_3$, and optionally comprises $Z_2O_5$, wherein X is a trivalent element, and Z is a pentavalent element,
wherein the zeolitic material contains from 3.8 to 12 wt.-% of Cu and/or Fe calculated as the respective element and based on 100 wt-% of $SiO_2$ contained in the zeolitic material having a CHA framework structure, and
wherein the $^{29}Si$ MAS NMR of the zeolitic material comprises:
a first peak (P1) in the range of from −96 to −98.8 ppm;
a second peak (P2) in the range of from −102 to −104.5 ppm; and
a third peak (P3) in the range of from −107.5 to −111 ppm;
wherein the integration of the first, second, and third peaks in the $^{29}Si$ MAS NMR of the zeolitic material offers a ratio of the integration values P1 : P2 : P3 ranging from (0.35 – 0.7) : 1 : (0.1 – 1.6).

11. The zeolitic material of claim 10, wherein X is selected from the group consisting of Al, B, In, Ga, and mixtures of two or more thereof.

12. The zeolitic material of claim 10 or 11, wherein the CHA framework comprises at least a portion of the Cu and/or Fe as extra-framework ions.

13. The zeolitic material of claim 12, wherein the molar ratio (Cu and/or Fe) : $X_2O_3$ of Cu and/or Fe to $X_2O_3$ of the framework structure ranges from 0.05 to 10.

14. Method for the treatment of $NO_x$ by selective catalytic reduction (SCR) comprising:
    (a) providing a catalyst comprising a zeolitic material according to any of claims 9 to 13; and
    (b) contacting a gas stream comprising $NO_x$ with the catalyst provided in (a).

15. Use of a synthetic organotemplate-free zeolitic material having a CHA framework structure according to any of claims 9 to 13 as a molecular sieve, as an adsorbent, for ion exchange, as a catalyst and/or as a catalyst support.

Figure 1a

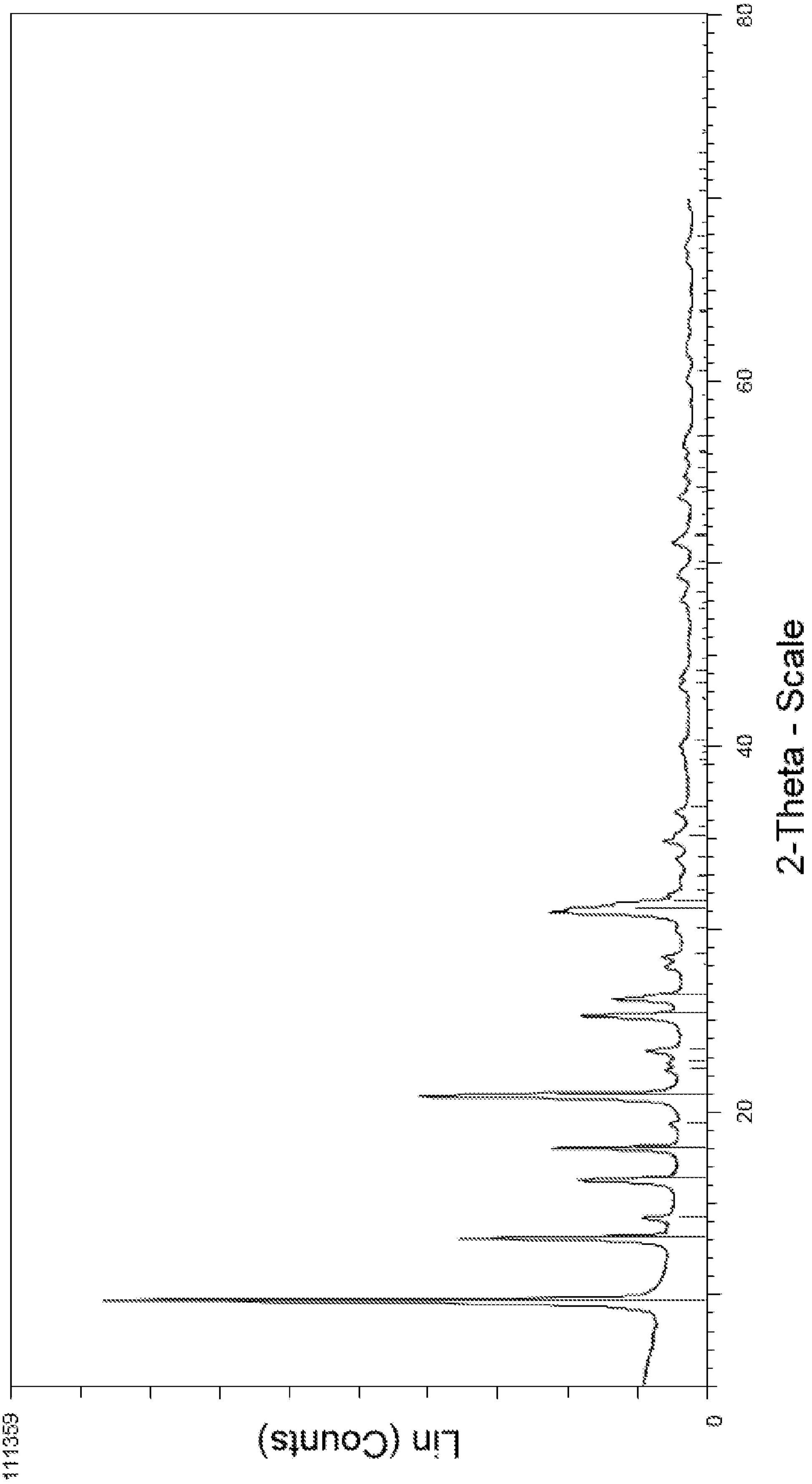
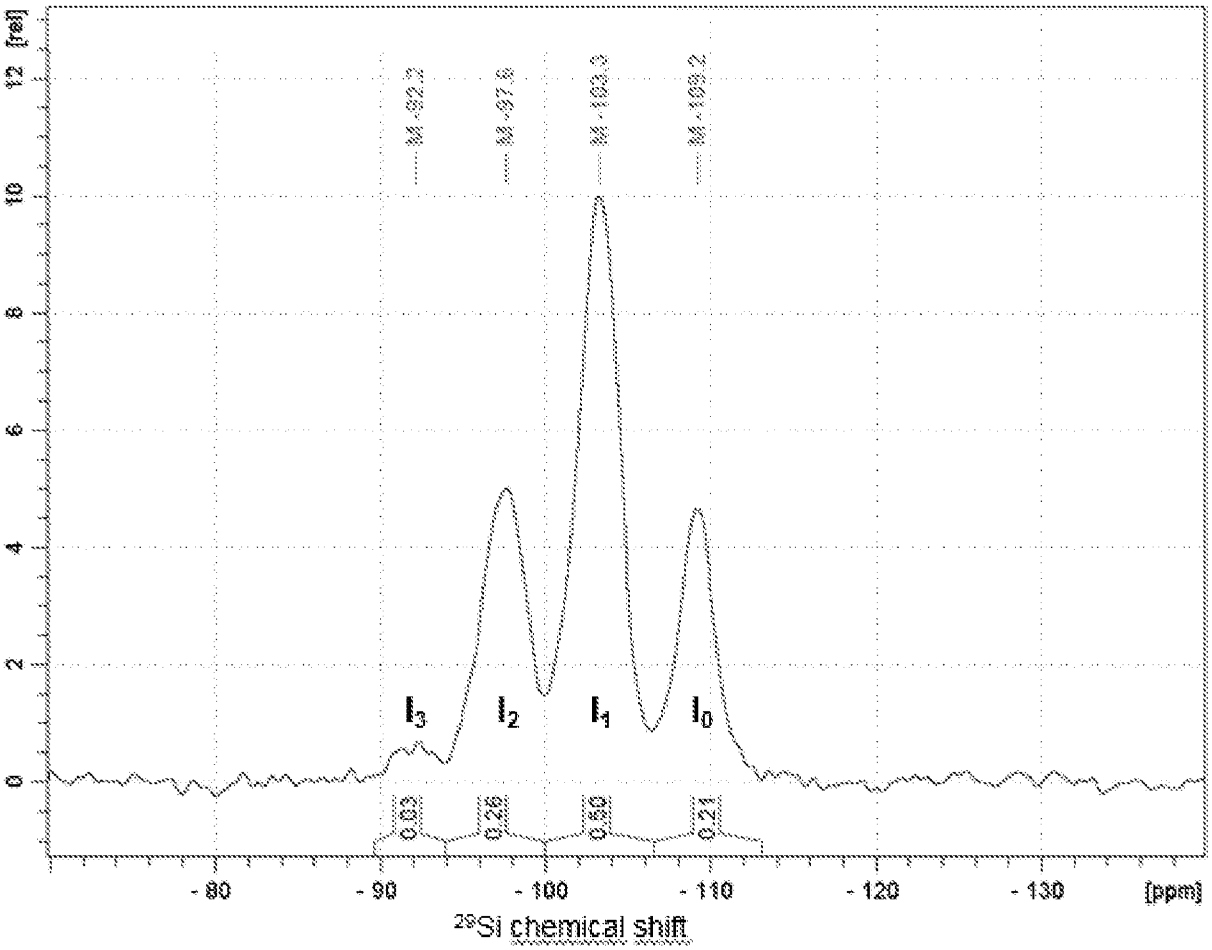


Figure 1b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8
6
4
2
0
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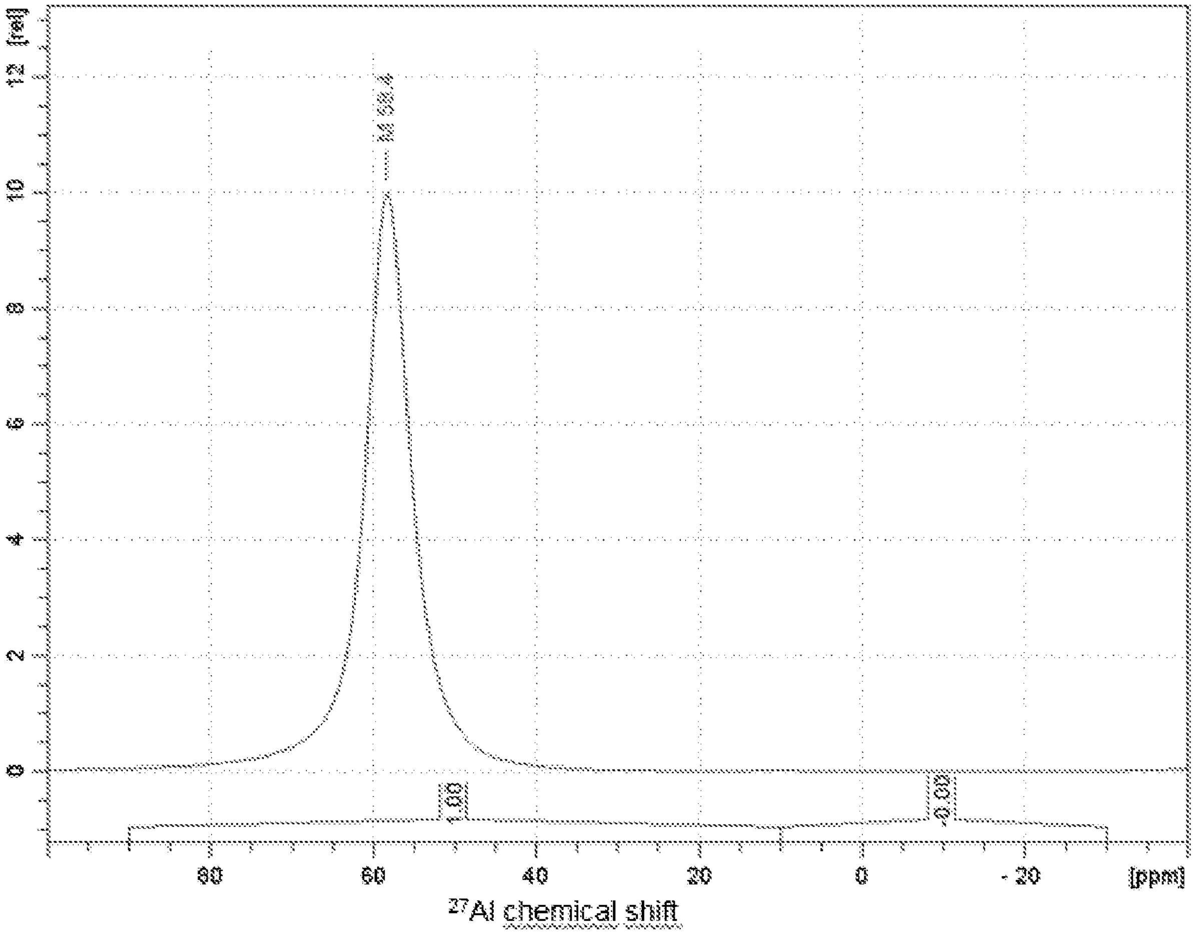
M -97.6
M -103.3
M -109.2
I3
I2
I1
I0
0.03
0.26
0.50
0.21
- 80
- 90
- 100
- 110
- 120
- 130
[ppm]
29Si chemical shift

Figure 1c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2
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60
40
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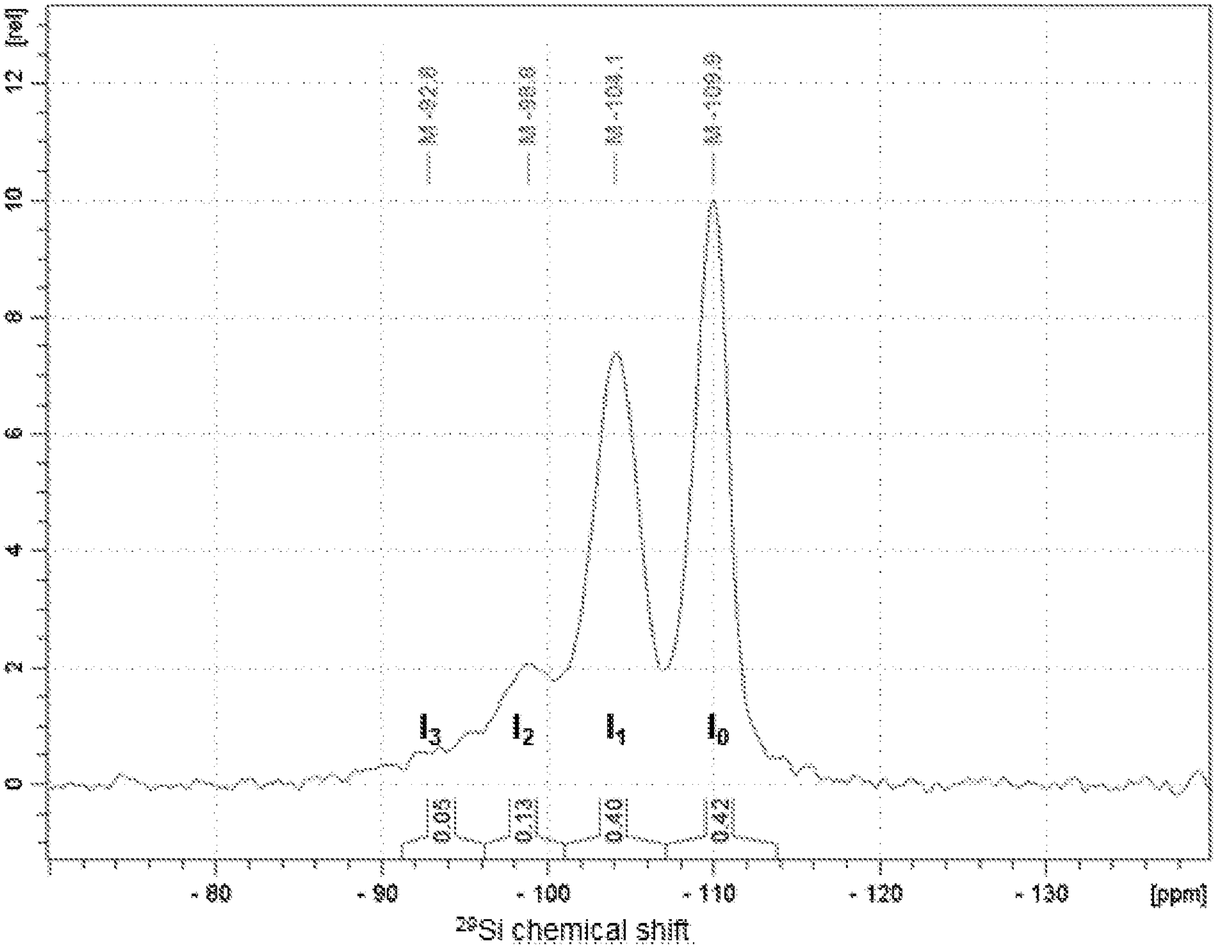
[ppm]
$^{27}$Al chemical shift

Figure 2a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6
4
2
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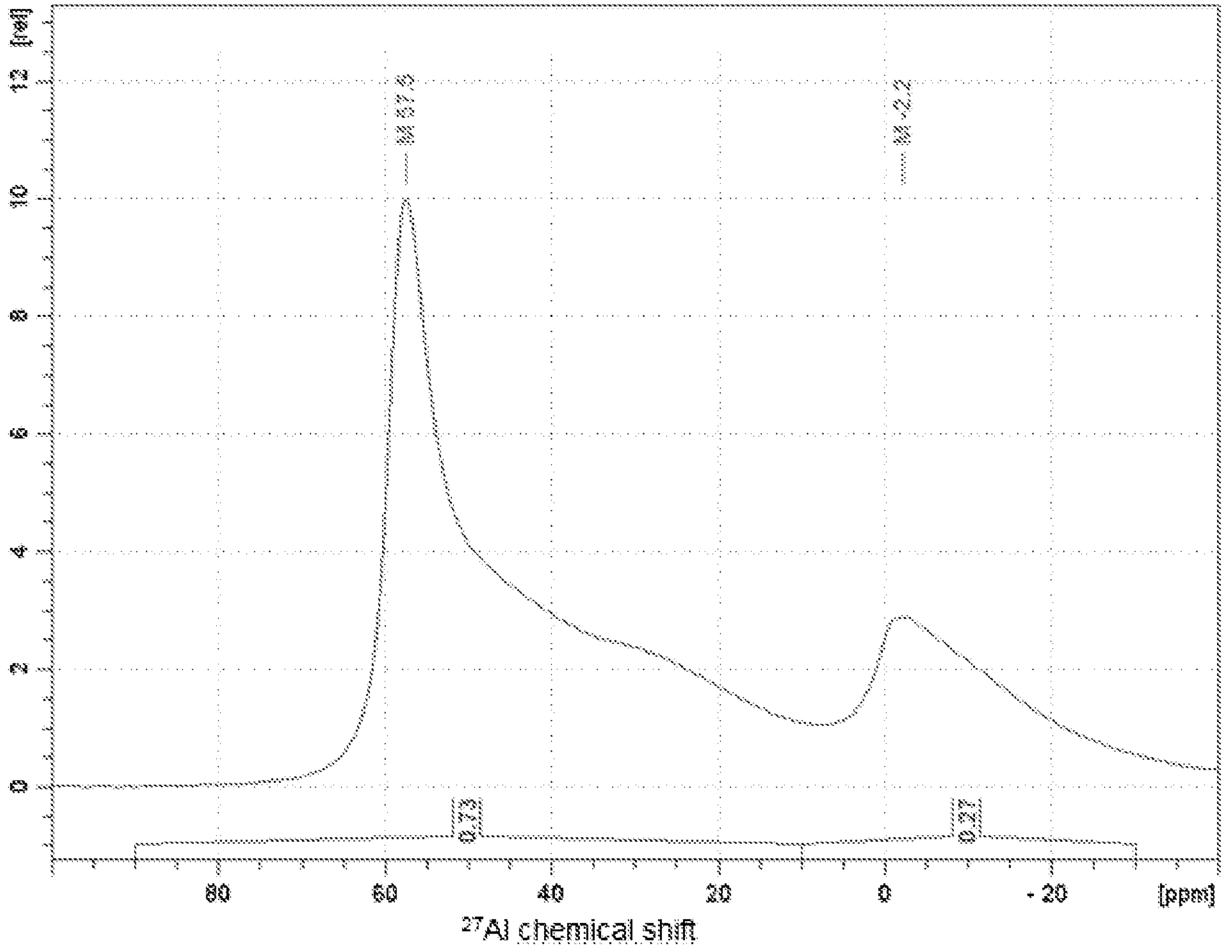
-104.1
-109.9
$I_3$
$I_2$
$I_1$
$I_0$
0.05
0.13
0.40
0.42
- 80
- 90
- 100
- 110
- 120
- 130
[ppm]
$^{29}$Si chemical shift

Figure 2b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0.27
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6
4
2
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80
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40
20
0
- 20
[ppm]
$^{27}$Al chemical shift

Figure 3

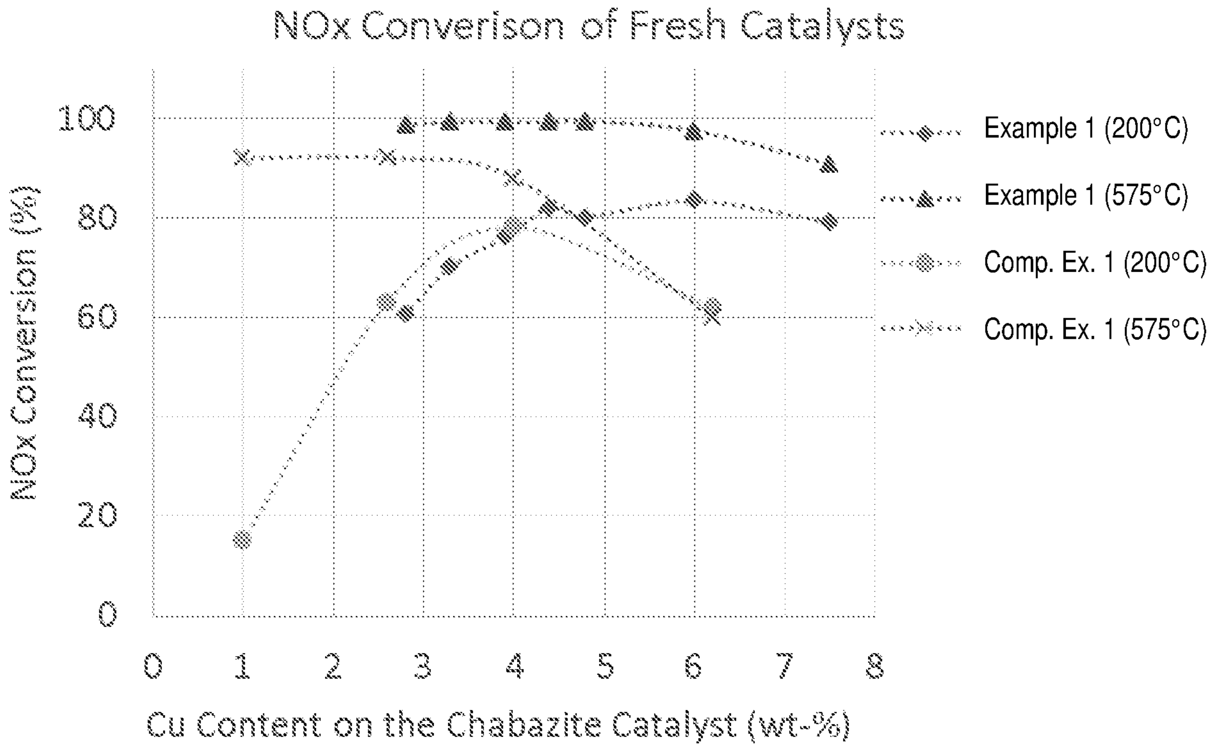
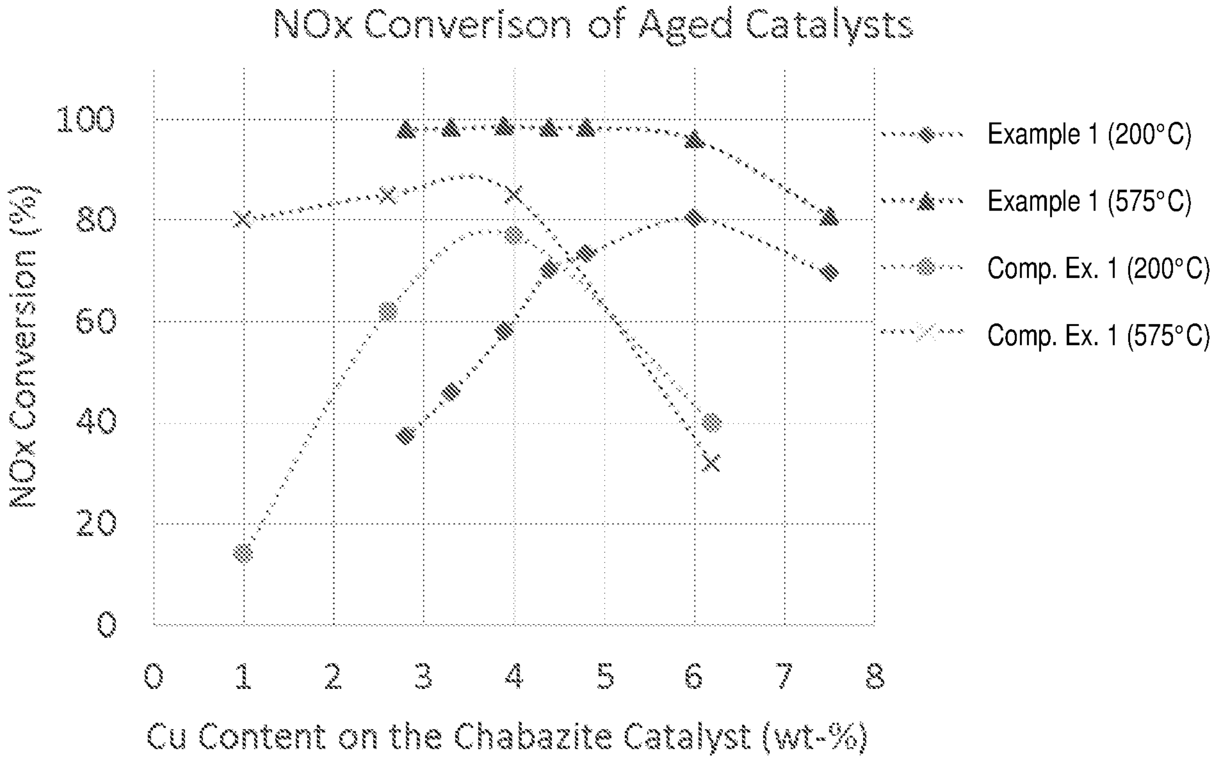

NOx Converison of Fresh Catalysts
NOx Conversion (%)
0
20
40
60
80
100
0
1
2
3
4
5
6
7
8
Cu Content on the Chabazite Catalyst (wt-%)
Example 1 (200°C)
Example 1 (575°C)
Comp. Ex. 1 (200°C)
Comp. Ex. 1 (575°C)

Figure 4


NOx Converison of Aged Catalysts
NOx Conversion (%)
0
20
40
60
80
100
0
1
2
3
4
5
6
7
8
Cu Content on the Chabazite Catalyst (wt-%)
Example 1 (200°C)
Example 1 (575°C)
Comp. Ex. 1 (200°C)
Comp. Ex. 1 (575°C)