

# Patent Owner's Demonstratives

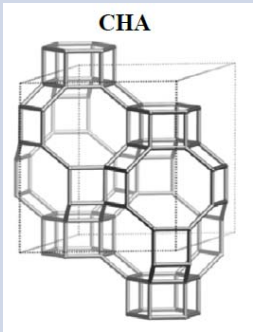
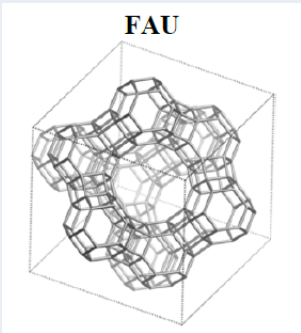
IPR2015-01121, -23, -24, -25

*Umicore AG & CO. KG v. BASF Corporation*  
(Oral Hearing – July 28, 2016)

# **Metal-Exchanged Zeolite Catalysts**

# Zeolite Overview

- Crystalline framework materials that contain pores of a molecular size.
- Framework types are cataloged by the IZA.

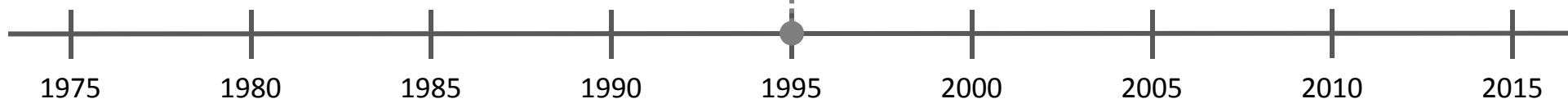
Framework	Materials	Pore Size	Ring Structure
 <p>CHA</p>	chabazite (natural) SSZ-13 SSZ-62	3.8 Angstroms	8-ring
 <p>FAU</p>	faujasite (natural) zeolite-Y	7.4 Angstroms	12-ring

Ex. 2018, ¶¶56-61

# Zeolite SCR Catalysts: Pre-662 Patent

“Indeed, several unresolved problems limit the outlook for successful use of zeolites in automotive converters: (i) hydrothermal stability, (ii) sensitivity to poisoning....A low hydrothermal stability, in particular, is the more critical weakness of copper-containing zeolites.”

Ex. 2012.005 (1995)

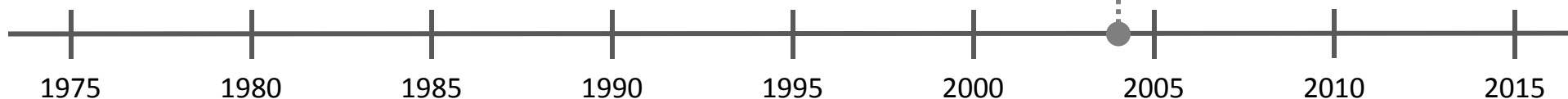


# Zeolite SCR Catalysts: Pre-662 Patent

“The catalyst was tested for durability in both wet and sulphur containing gases, but even water had a negative long-term effect on the catalyst and its applicability must be regarded as rather low. One can be quite pessimistic about the use of zeolites for several reasons:

- (i) The catalyst types have been tested in the SCR reaction since the eighties without a commercial breakthrough.
- (ii) Zeolites have been extensively tested in the so-called HC-SCR reaction with insufficiently long-term stability....

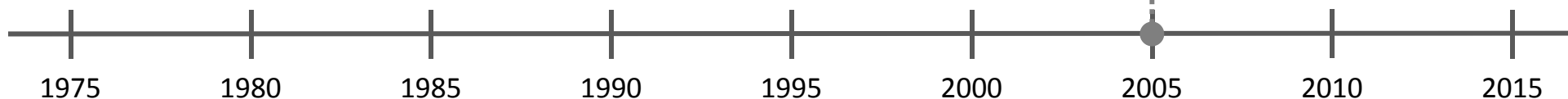
Ex. 2026.006 (2004)



# Zeolite SCR Catalysts: Pre-662 Patent

“The aim of this study was to obtain knowledge about the activity of zeolite-based catalysts in the NH<sub>3</sub>-SCR reaction with excess of oxygen. The limited hydrothermal stability of zeolites may restrict their use.”

Ex. 2021.002 (2004)

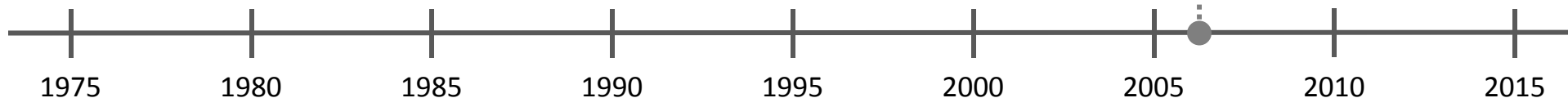


# Zeolite SCR Catalysts: Pre-662 Patent

“Consequently, achieving hydrothermal stability of the catalyst is a critical issue in the commercial application of urea SCR technology to the exhaust stream from diesel engines.”

“CuZSM5 has been reported as one of the most promising catalysts for the SCR of NO<sub>x</sub> from light- and heavy- duty diesel engines with urea. However, the hydrothermal stability of the catalyst is another critical issue to be resolved for the commercial application of urea SCR technology to automotive engines. CuZSM5 after hydrothermal aging under simulated flue gas stream at temperature above 600° C with 10% water reveals significant catalyst deactivation.

Ex.2024.001, .010 (2006)

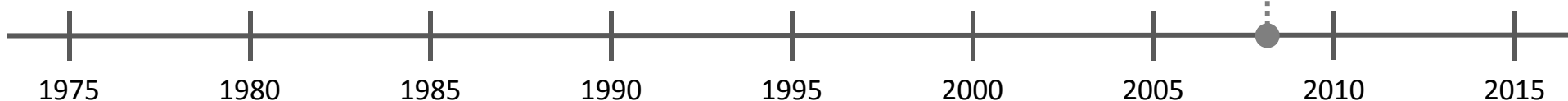


# Zeolite SCR Catalysts: Pre-662 Patent

“Although metal-exchanged zeolites have proven to be very active SCR catalysts, using these materials in exhaust after-treatment systems on diesel vehicles is a difficult task. One of the challenges for the practical application of SCR catalysts is their durability under hydrothermal conditions. Fe-ZSM-5 catalysts, for instance, have been reported to be very stable even in the presence of H<sub>2</sub>O, but at temperatures above 500° C deactivation is always observed.”

“One of the major requirements for the practical application of zeolites in the SCR process is their durability under hydrothermal conditions, which is not yet sufficient. There is no guarantee that a zeolite-based catalyst will be found that meets all of the diverse requirements for future SCR systems on diesel vehicles. Research is now targeting catalysts that combine both high activity at low temperatures and hydrothermal stability at high temperatures.”

Ex. 2022.016, .029 (2008)



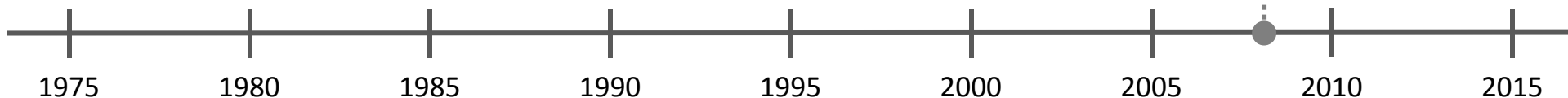


# Zeolite SCR Catalysts: Post-662 Patent

“Improvements in the thermal durability of Cu/zeolite based SCR formulations has been highly desirable and pursued by many research institutes and catalyst suppliers.”

“In this work, a laboratory flow reactor was utilized to hydrothermally age and evaluate the latest state-of-the-art Cu/zeolite formulations. Results confirm remarkable high temperature stability up to 950 °C while maintaining stable low temperature NO<sub>x</sub> activity. A broad range of time-at-temperature hydrothermal aging was carried out to clearly define the full durability range. The aging time was varied from 1 hour to 256 hours while aging temperature was varied from 670 °C to 1100 °C.”

Ex. 2002.001-.002 (2008)

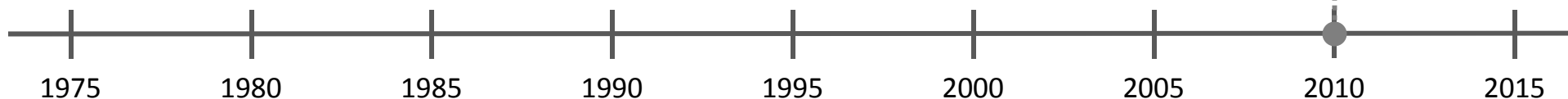


# Zeolite SCR Catalysts: Post-662 Patent

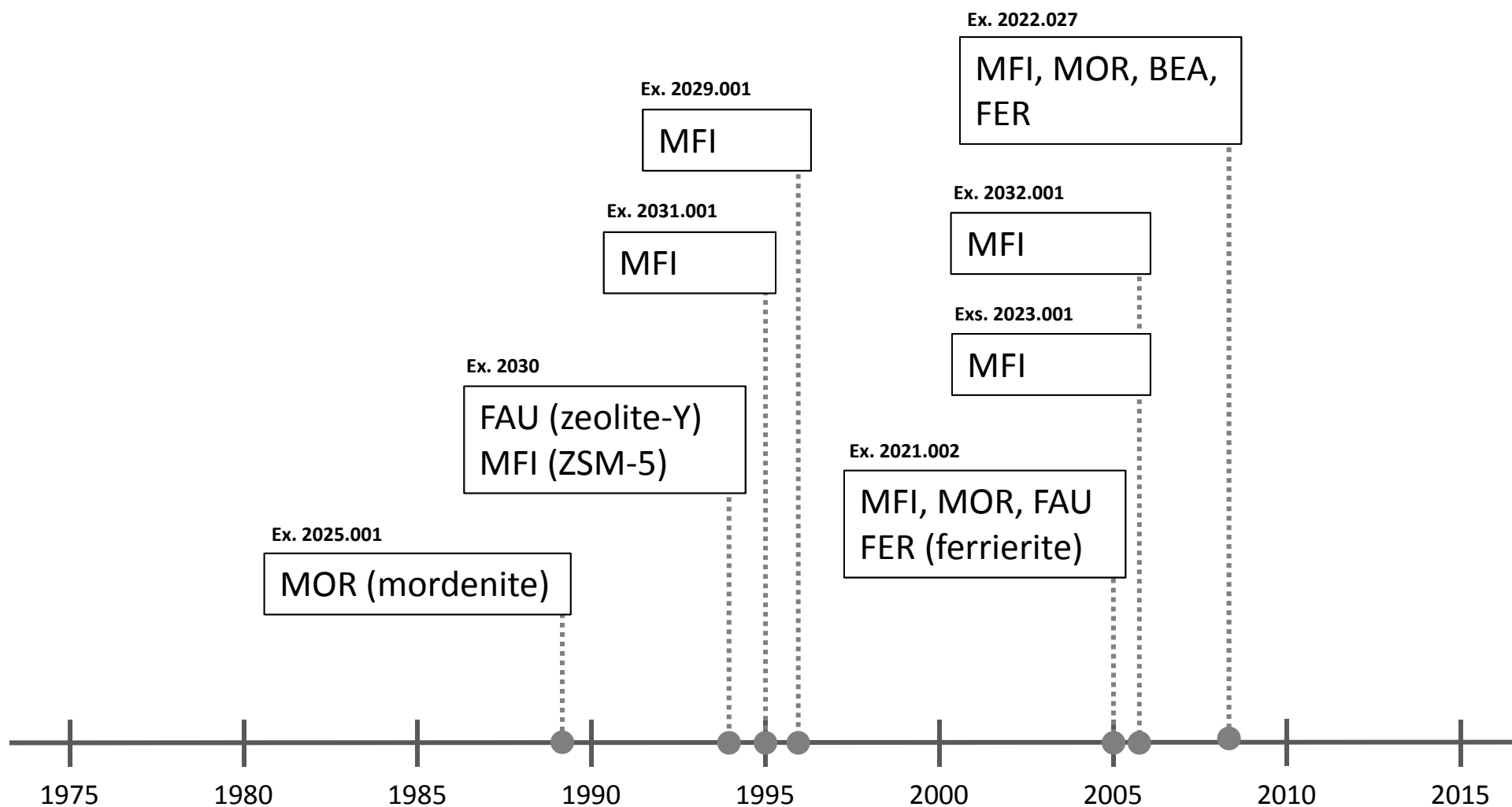
“In the very recent patent literature, Cu<sup>2+</sup> ion-exchanged SSZ-13 (Cu-SSZ-13) has been reported to exhibit NO<sub>x</sub> conversions of 90-100% over a wide temperature range in the NH<sub>3</sub>-SCR process, and its activity exceeded 80% even after extensive high-temperature hydrothermal aging [9]. The SSZ-13 has chabazite (CHA) structure with a relatively small pore radius (~3.8 Å) in an eight-membered ring [10].”

“Our results confirm that the activity and selectivity of the Cu-SSZ-13 catalyst for both NO<sub>x</sub> SCR with NH<sub>3</sub> and NH<sub>3</sub> oxidation are superior to those of both Cu-beta and Cu-ZSM-5.”

Ex. 2014.001 (2010)



# Non-CHA Framework Types: Pre-662 Patent



# CHA Framework Type: Pre-662 Patent

MOR  
ERI  
NAT  
CHA  
FAU

Suitable natural zeolites are:

Mordenite:  $(\text{Ca}, \text{K}_2, \text{Na}_2) [\text{AlSi}_5\text{O}_{12}]_2 \cdot 7\text{H}_2\text{O}$

Erionite:  $(\text{K}_2, \text{Na}_2, \text{Ca}) [\text{AlSi}_3\text{O}_8]_2 \cdot 6\text{H}_2\text{O}$

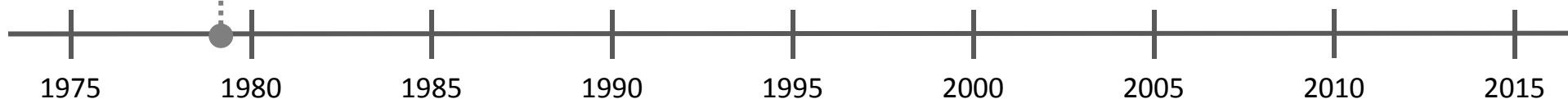
Natrolite:  $\text{Na}_2[\text{Al}_2\text{Si}_3\text{O}_{10}] \cdot 2\text{H}_2\text{O}$

Chabazite:  $(\text{Ca}, \text{Na}_2) [\text{Al}_2\text{Si}_4\text{O}_{12}] \cdot 6\text{H}_2\text{O}$

Faujasite:  $\text{Na}_2\text{Ca} [\text{Al}_2\text{Si}_4\text{O}_{12}]_2 \cdot 16\text{H}_2\text{O}$

Especially preferred crystalline aluminosilicates are those having a pore diameter in the range of about 6–13 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 2–6. For exam-

Ex. 1002 at 4:6-12, 36-38



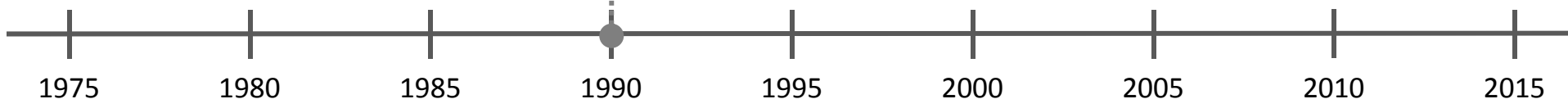
# CHA Framework Type: Pre-662 Patent

“...it appears that SO<sub>2</sub> poisoning has both short term and long term effects. For example, flowing a gas stream containing 2,000 parts per million by volume ("Vppm") SO<sub>2</sub> through catalysts comprising copper-promoted small to medium pore zeolites such as...naturally occurring chabazite...resulted in 10 to 40 percent reduction in SCR process activity. Even at SO<sub>2</sub> levels as low as 130 Vppm SO<sub>2</sub>, significant activity reduction for the SCR process was noted for such catalysts.”

“A 60 percent reduction in SCR process activity is typical for Fe<sub>2</sub>O<sub>3</sub> containing natural chabazite.”

“It has been found that zeolites which are highly resistant to sulfate poisoning and provide good activity....are zeolites which have pores which exhibit a pore diameter of at least about 7 Angstroms and are interconnected in three dimensions.”

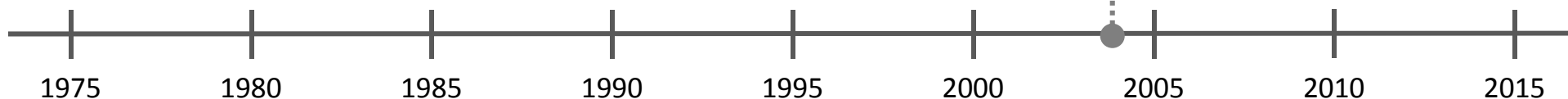
Ex. 1010 at 4:62-68, 5:23-25 6:12-20



# CHA Framework Type: Pre-662 Patent

“Also provided by the present invention is an improved process for the reduction of oxides of nitrogen contained in a gas stream in the presence of oxygen wherein said process comprises contacting the gas stream with a zeolite, the improvement comprising using as the zeolite a zeolite having the CHA crystal structure, a mole ratio greater than about 10 of silicon oxide to aluminum oxide and having a crystallite size of 0.5 micron or less. The zeolite may contain a metal or metal ions (such as cobalt, copper or mixtures thereof) capable of catalyzing the reduction of the oxides of nitrogen, and may be conducted in the presence of a stoichiometric excess of oxygen. In a preferred embodiment, the gas stream is the exhaust stream of an internal combustion engine.”

Ex. 1004 at 1:54-67



# CHA Framework Type: Post-662 Patent

Ex. 2002

Cavataio, G., et al. *“Enhanced Durability of a Cu/Zelite Based SCR Catalyst,”* SAR Int. J. Fuels. Lubr., Vol. 1, Issue 1.

Ex. 2014

Kwak, J., et al., *“Excellent Activity and Selectivity of Cu-SSZ-13 in the Selective Catalytic Reduction of NO<sub>x</sub> with NH<sub>3</sub>,”* Journal of Catalysis.

Ex. 2020

Gao F., et al., *“Effects of Si/Al ratio on Cu/SSZ-13 NH<sub>3</sub>-SCR catalysts: Implications for the active species and the roles of Bronsted acidity,”* Journal of Catalysis.



## '662 and '203 Patents



# '662 Claims

- **Claim 1 (catalyst):**
  - aluminosilicate CHA
  - SAR: 15-150
  - Cu/Al: 0.25-1
  - NH<sub>3</sub>-SCR of NO<sub>x</sub>
- **Dep. Claims 2-8:**
  - SAR: 15-100, 25-40, 30
  - Cu/Al: 0.30-0.50, 0.40
- **Dep. Claims 12-14, 32, 39, 40**
  - Catalyst deposited on substrate
- **Dep. Claims 15-20, 41-46**
  - Substrate coated with CuCHA, or coated with Pt and CuCHA
- **Dep. Claims 21-24, 47-50**
  - Catalyst disposed downstream of a diesel engine
- **Dep. Claims 33-38**
  - Exhaust gas treatment system including catalyzed soot filter and diesel oxidation catalyst located upstream of the CuCHA catalyst

# Properties of the Claimed CuCHA Catalyst

## Example 1 of '662 Patent

SAR = 30, Cu/Al = 0.30

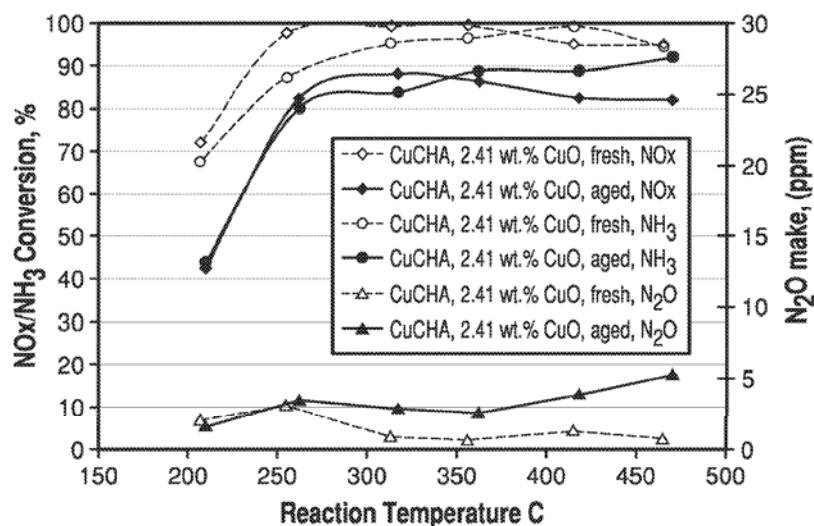


FIG. 1

## Example 2 of '662 Patent

SAR = 30, Cu/Al = 0.33

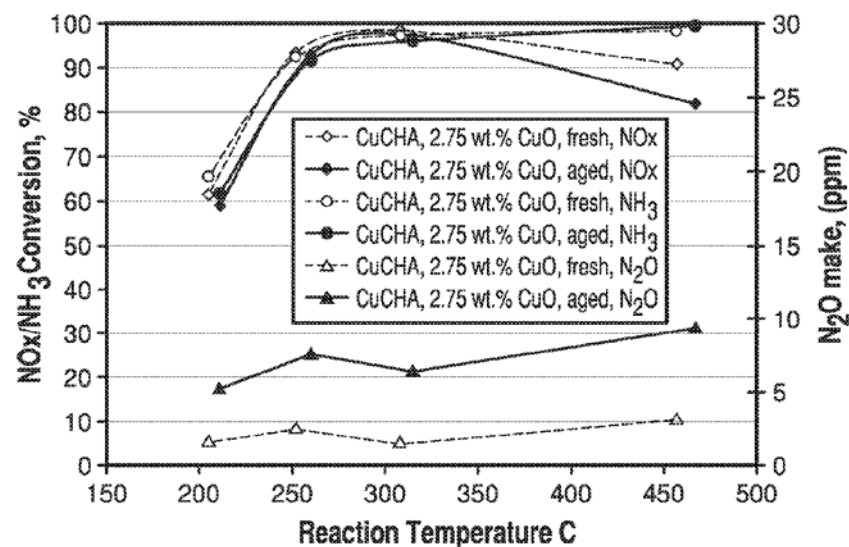
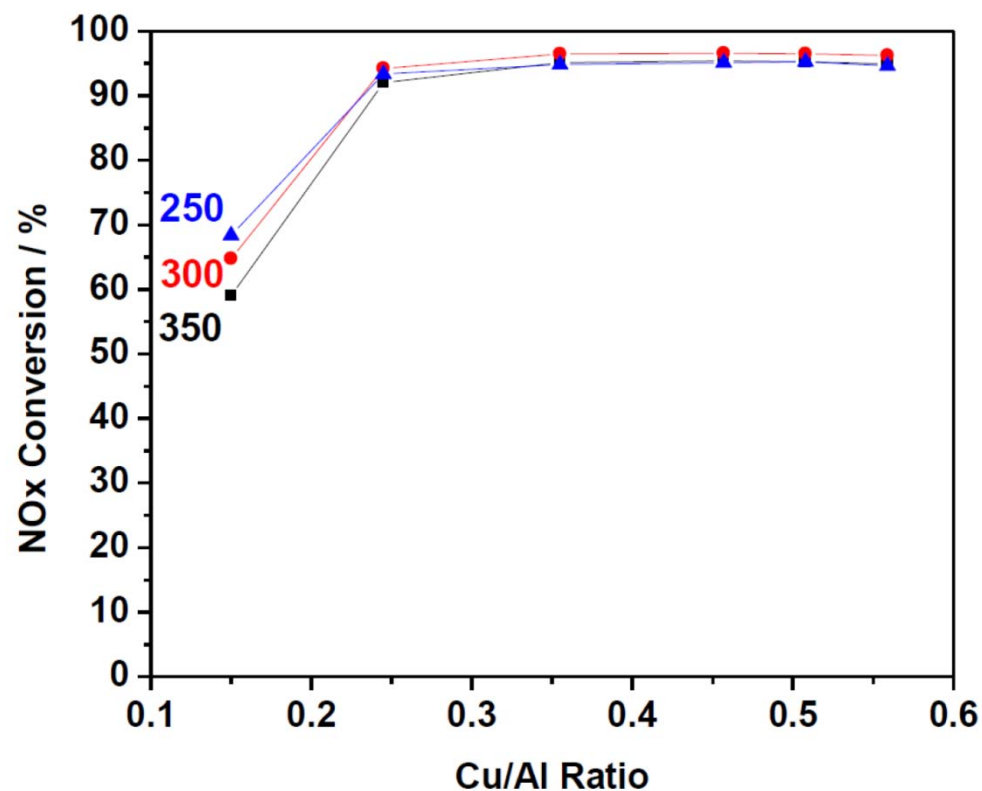


FIG. 2

# Criticality of Claimed Ranges: Cu/Al

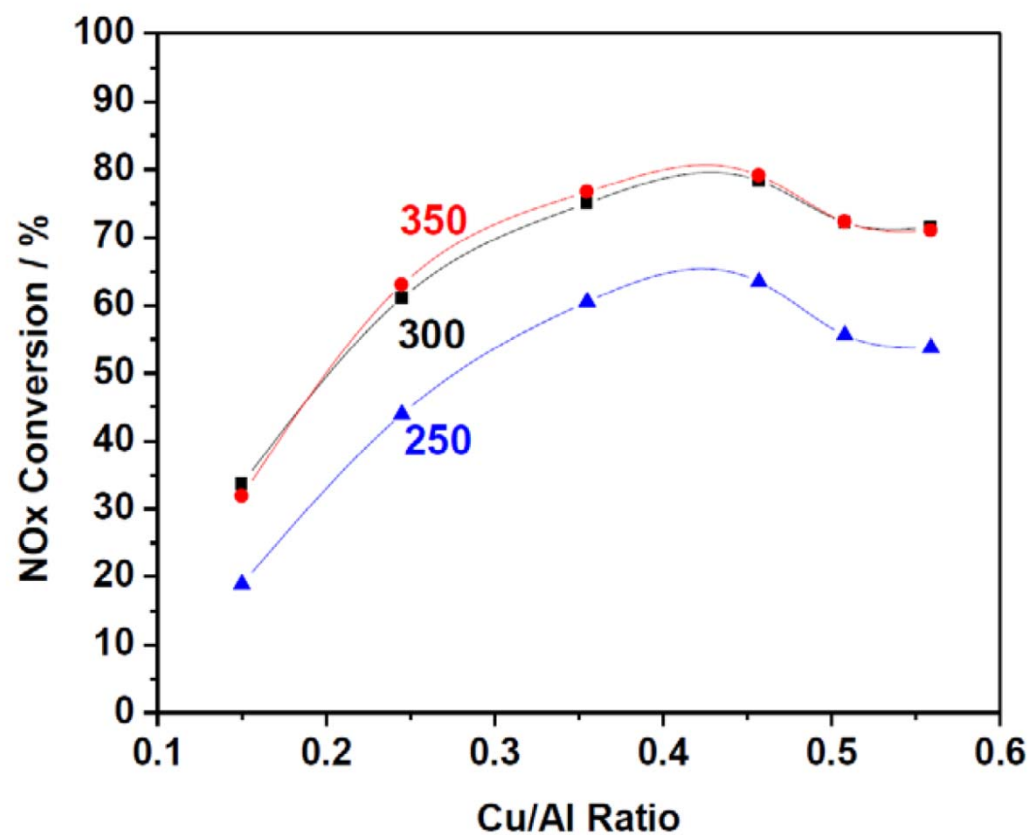
Fresh Cu-CHA, SAR 30, Cu/Al from 0.15 to 0.6



Ex. 2018 at ¶149

# Criticality of Claimed Ranges: Cu/Al

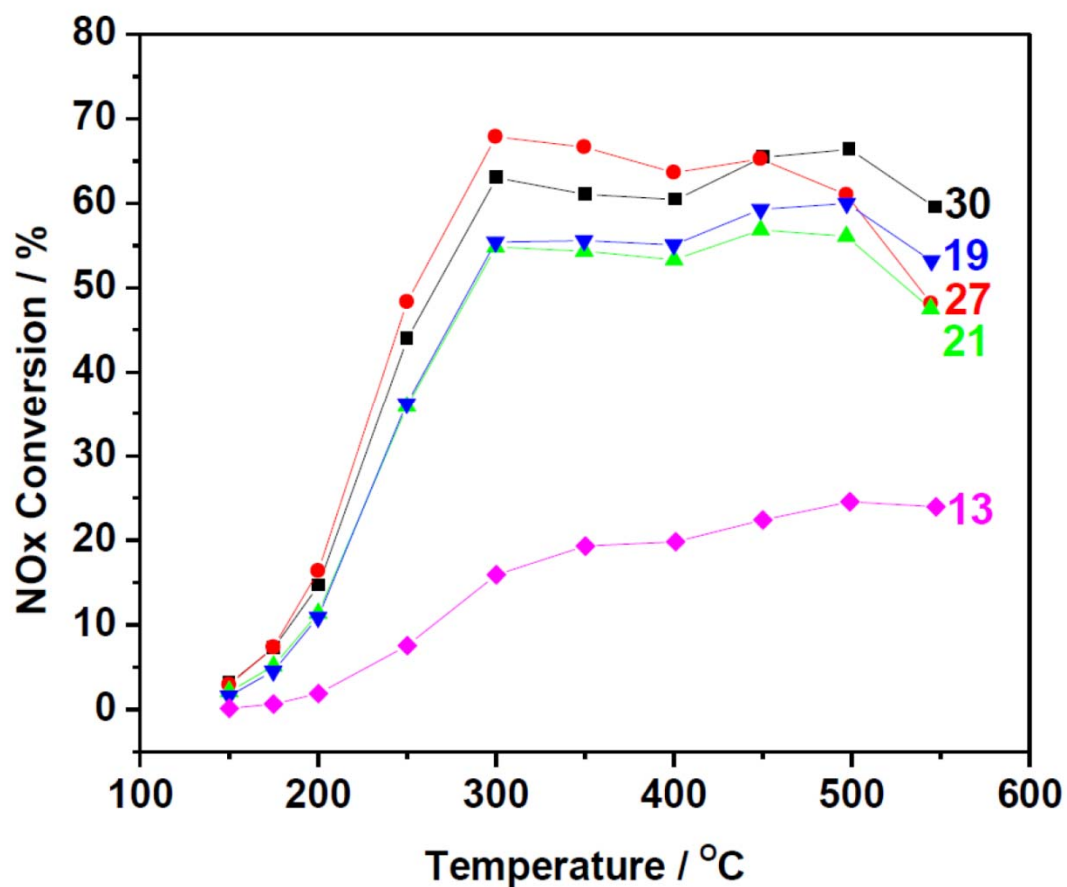
Aged Cu-CHA, SAR 30, Cu/Al from 0.15 to 0.6



Ex. 2018 at ¶148

# Criticality of Claimed Ranges: SAR

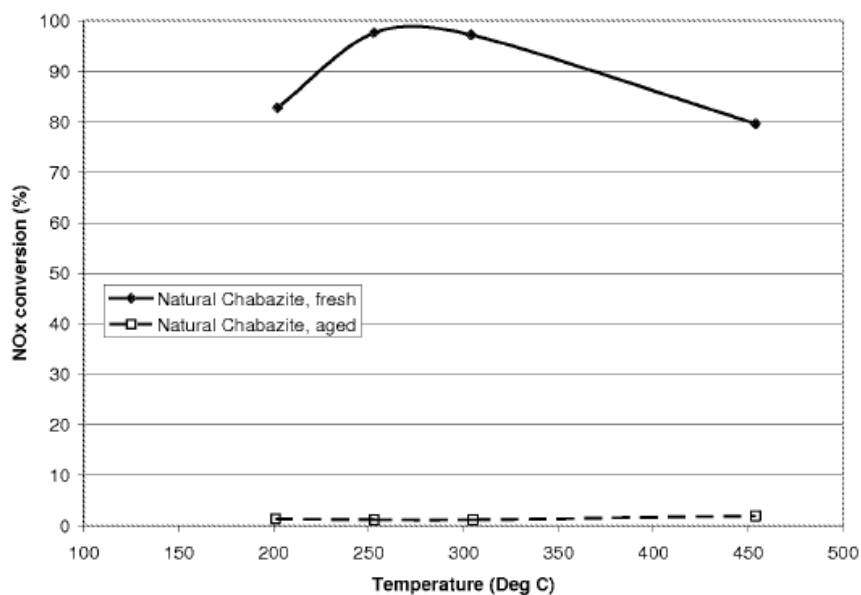
Aged Cu-CHA, Varying SAR, Cu/Al~0.245



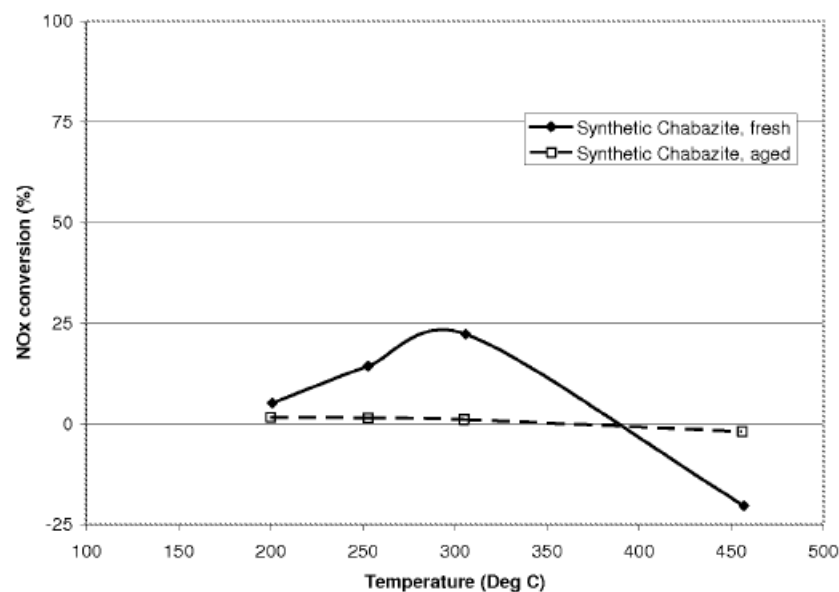
Ex. 2018 at ¶150

# Properties of Prior Art CuCHA

**Natural Chabazite**  
**SAR 6.3, Cu/Al 0.32**

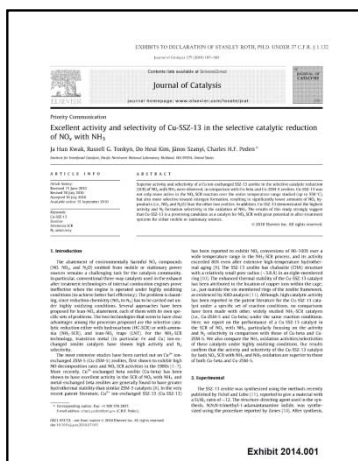


**Synthetic Chabazite**  
**SAR 4.5, Cu/Al 0.33**



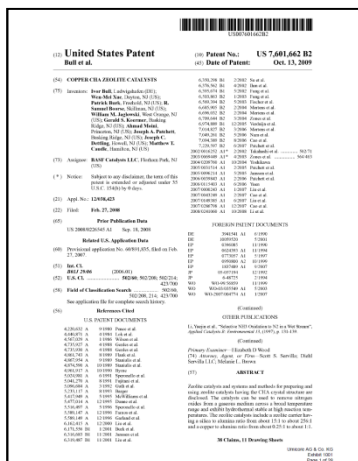
Ex. 2011 at ¶¶11-14

# Properties of Prior Art Cu-Y, Cu-Beta, and Cu-ZSM



“Our results confirm that the activity and selectivity of the Cu-SSZ-13 catalyst for both NO<sub>x</sub> SCR with NH<sub>3</sub> and NH<sub>3</sub> oxidation are superior to those of both Cu-beta and Cu-ZSM-5.”

Ex. 2014.001 (2010)



Cu-Y  
Cu-Beta

TABLE 1

Example	Cu/Al		NO <sub>x</sub> conversion (%)				N <sub>2</sub> O make, ppm	
	Atomic ratio	CuO %	210° C., fresh	210° C., aged	460° C., fresh	460° C., aged	460° C., fresh	460° C., aged
1	0.30	2.41	75	43	95	82	0.8	5.3
2	0.33	2.75	62	59	90	83	3.1	9.3
3	0.38	3.36	74	70	91	81	2.7	10.5
4	0.44	3.85	76	60	88	72	3.5	14.2
5	0.24	1.94	50	30	95	75	0.2	5.0
10	0.23	4.6	43	42	99	96	26	51
11	0.36	2.5	92	23	84	53	10	9.4
12	0.46	3.7	75	78	89	80	5.4	11.7
1A	0.40	3.2		61		82		11.3

## **Zones in view of Maeshima**

('662 Claims 1-8, 30)

('203 Claims 1, 14, 15, 17-22, 26, 27)



# Zones '644: Small Crystal Size CHA Zeolite

## FIELD OF THE INVENTION

The present invention relates to crystalline zeolite SSZ-62 that has the CHA crystal structure, a mole ratio greater than 10 of silicon oxide to aluminum oxide and has a crystallite size of 0.5 micron or less. The present invention also relates to a method for preparing SSZ-62 using specific sources of silicon and aluminum, and a N,N,N-trimethyl-1-adamantammonium cation templating agent, to processes employing SSZ-62 as a catalyst, and to processes using SSZ-62 to separate gasses.

Ex.1004 at 1:5-15

# Zones '644: General Disclosure Regarding Processes

“converting lower alcohols and other oxygenated hydrocarbons”

Ex.1004 at 1:46-53, 5:17-35, 7:4-23

“catalyzing the reduction of oxides of nitrogen, and may be conducted in the presence of a stoichiometric excess of oxygen”

Ex.1004 at 1:61-65

“as a catalyst to prepare dimethylamine”

Ex.1004 at 5:36-64

“SSZ-62 can also be used to separate gasses”

Ex.1004 at 5:66-6:2

# Zones '644: General Disclosure Regarding Processes

“Also provided by the present invention is an improved process for the reduction of oxides of nitrogen contained in a gas stream in the presence of oxygen wherein said process comprises contacting the gas stream with a zeolite, the improvement comprising using as the zeolite a zeolite having the CHA crystal structure, a mole ratio greater than about 10 of silicon oxide to aluminum oxide and having a crystallite size of 0.5 micron or less. The zeolite may contain a metal or metal ions (such as cobalt, copper or mixtures thereof) capable of catalyzing the reduction of the oxides of nitrogen, and may be conducted in the presence of a stoichiometric excess of oxygen. In a preferred embodiment, the gas stream is the exhaust stream of an internal combustion engine.”

Ex.1004 at 1:54-67

- Reduction of NO without reducing agents
- Decomposition of NO in presence of oxygen
- N<sub>2</sub>O decomposition
- NH<sub>3</sub>-SCR of NO<sub>x</sub>
- HCR-SCR of NO<sub>x</sub>

Ex.2009 at ¶18

Ex.2018 at ¶79

Ex.2027 at 37:19-38:22

## Zones '644: No Disclosure of Using SSZ-62 for NH<sub>3</sub>-SCR of NO<sub>x</sub>

APPLICATION NO.		FILING DATE	FIRST NAMED INVENTOR	ATTORNEY DOCKET NO.	CONFIRMATION NO.
95081,453		09/28/2010	7,801,662	04/19/000100.36	2755

13172 7390 06/14/2012  
Diehl Servilla LLC (COG/COG)  
33 Wood Avenue South  
Second Floor, Suite 210  
Iselin, NJ 08830

Please find below and/or attached an  
The time period for reply, if any, is set

While Zones teaches at col. 1, lines 61-65 that its CHA zeolite, such as SSZ-62, may contain a metal ion, such as copper, "capable of catalyzing the reduction of oxides of nitrogen", no amount is ever exemplified for such catalysis, and **Zones does not teach using its catalyst for ammonia SCR of NO<sub>x</sub>.** As noted by Patent Owner on p. 29 of the Remarks filed 12/19/11 :

PTOL-90A (Rev. 04/07) Exhibit 2006.001

*Ex. 2006.026-.027*

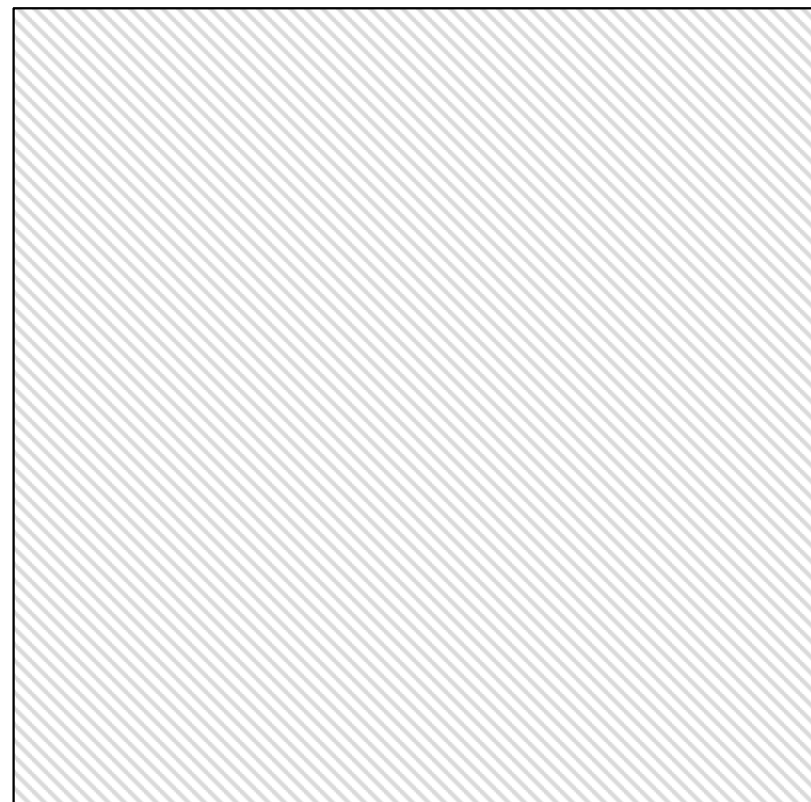
## Zones '644: No Disclosure of the Properties of SSZ-62 for the SCR of NO<sub>x</sub>

### Converting Methanol

“The catalyst gives greater than 90% selectivity for C<sub>2</sub>-C<sub>4</sub> olefins and does not show methanol breakthrough for greater than 15 hours. Prior art catalysts with larger crystallite sizes than SSZ-62 (like SSZ-13 with a crystallite size of about 1.2 microns and a silica/alumina mole ratio of about 9 or 18) show breakthrough at about 5 hours on stream under these conditions. The smaller crystallite SSZ-62 gives superior performance in this application.”

Ex.1004 at 7:16-23

### Reduction of NO<sub>x</sub>



Ex.1004

# Zones '644 Compared to Byrne

## Byrne

“...copper-promoted small to medium pore zeolites such...naturally occurring chabazite...resulted in 10 to 40 percent reduction in SCR process activity. Even at SO<sub>2</sub> levels as low as 130 Vppm SO<sub>2</sub>, significant activity reduction for the SCR process was noted for such catalysts. On the other hand, larger pore zeolites such as Y, L and USY exhibited no short-term SO<sub>2</sub> susceptibility....A 60 percent reduction in SCR process activity is typical for Fe<sub>2</sub>O<sub>3</sub> containing natural chabazite.”

Ex.1010 at 4:65-5:26

## Zones '644

“...a zeolite having the CHA crystal structure, a mole ratio greater than about 10 of silicon oxide to aluminum oxide and having a crystallite size of 0.5 micron or less. The zeolite may contain a metal or metal ions (such as cobalt, copper or mixtures thereof) capable of catalyzing the reduction of the oxides of nitrogen, and may be conducted in the presence of a stoichiometric excess of oxygen.”

Ex.1004 at 1:61-65

# Maeshima: $\text{NH}_3$ -SCR of $\text{NO}_x$ in Stationary Source

In short, the present invention relates to a method for selectively reducing nitrogen oxides contained in exhaust gases from stationary sources, such as flue gas from the combustion furnace of power plants, by using ammonia as a reducing agent, and the most characteristic feature of the present invention resides in that the catalyst used is contacted with ammonia in an amount excessive over the stoichiometric amount necessary for reduction of nitrogen oxides in an exhaust gas to thereby activate the catalyst and then, the amount of ammonia is reduced to a minimum amount necessary for reduction of the nitrogen oxides to thereby effect the catalytic reduction.

Ex.1002 at 2:9-21



# Maeshima: Temperature Limitations

lyst. As the reaction conditions, there may be adopted a reaction temperature of about 200° to about 500° C., preferably about 250° to about 400° C., and a gas space velocity of about 2,000 to about 100,000 V/H/V, preferably about 5,000 to about 30,000 V/H/V. Since the activity of the ammonia reduction of nitrogen oxides is lowered at higher or lower temperatures, good results are obtained when a mixture of the exhaust gas and ammonia is contacted with the catalyst bed at a temperature within the above-mentioned range.

Ex.1002 at 3:23-32



# Maeshima: Zeolite Frameworks

Suitable natural zeolites are:

Mordenite:  $(\text{Ca}, \text{K}_2, \text{Na}_2) [\text{AlSi}_5\text{O}_{12}]_2 \cdot 7\text{H}_2\text{O}$

Erionite:  $(\text{K}_2, \text{Na}_2, \text{Ca}) [\text{AlSi}_3\text{O}_8]_2 \cdot 6\text{H}_2\text{O}$

Natrolite:  $\text{Na}_2[\text{Al}_2\text{Si}_3\text{O}_{10}] \cdot 2\text{H}_2\text{O}$

Chabazite:  $(\text{Ca}, \text{Na}_2) [\text{Al}_2\text{Si}_4\text{O}_{12}] \cdot 6\text{H}_2\text{O}$

Faujasite:  $\text{Na}_2\text{Ca} [\text{Al}_2\text{Si}_4\text{O}_{12}]_2 \cdot 16\text{H}_2\text{O}$

The natural zeolites contain alkaline earth metals, with alkali metals.

As the synthetic zeolites, there may be used synthetic faujasite and synthetic mordenite. The synthetic faujasites include:

Zeolite-A:  $1.0 \pm 0.2\text{M}_{2/n}\text{O} : \text{Al}_2\text{O}_3 : 1.85 \pm 0.5\text{SiO}_2 : \text{YH}_2\text{O}$  (wherein M represents a metal,  $n$  represents a valence of M and Y represents a number of about 6 or below).

Zeolite-X:  $1.0 \pm 2\text{M}_{2/n}\text{O} : \text{Al}_2\text{O}_3 : 5 \pm 0.5\text{SiO}_2 : \text{YH}_2\text{O}$  (wherein M represents a metal of a valence of from 1 to 3, inclusive,  $n$  represents a valence of M and Y represents a number of about 8 or below).

Zeolite-Y:  $0.9 \pm 0.2\text{Na}_2\text{O} : \text{Al}_2\text{O}_3 : \text{WSiO}_2 : \text{YH}_2\text{O}$  (wherein W represents a number between 3 and 6, inclusive, and Y represents a number of about 9 or below).

The synthetic mordenites include, for example:

Zeolite-L:  $1.0 \pm 0.1\text{M}_{2/n}\text{O} : \text{Al}_2\text{O}_3 : 0.4 \pm 0.5\text{SiO}_2 : \text{YH}_2\text{O}$  (wherein M represents a metal,  $n$  represents a valence of M, and Y represents a number from 0 to 7, inclusive).

Ex.1002 at 4:6-35

## Maeshima: Use Low SAR, Large Pore Size Zeolite Frameworks

Especially preferred crystalline aluminosilicates are those having a pore diameter in the range of about 6–13 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 2–6. For example, synthetic faujasite having a pore diameter of about 8–9 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 2–3 and other synthetic faujasites having a pore diameter of about 8–9 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 4–6 are preferred.

Ex.1002 at 4:36-43

# Maeshima: Metal Ions

The above-mentioned active metal component having an activity of reducing nitrogen oxides is supported on the thus prepared aluminosilicate carrier by the impregnation treatment. Most preferred metals are copper, iron, chromium, and vanadium, and better results are obtained when at least one metal selected from them is employed.

Ex.1002 at 6:1-7

# Maeshima: Ion Exchange Ratio Not Critical

A zeolite catalyst having incorporated therein an active metal ion is prepared by contacting a crystalline aluminosilicate with an aqueous or organic solution of an active metal compound according to a customary method. The ion exchange ratio is not particularly critical, but it is generally preferred that the ion exchange ratio be about 60 to about 100%.

Ex.1002 at 4:44-50

# Petitioner Assumes That CuCHA Is Used For NH<sub>3</sub>-SCR of NO<sub>x</sub>

152. While Zones does not expressly reference catalysts with Cu/Al ratios within the claimed ranges, it does recognize that “the zeolite may contain a metal or metal ions (such as . . . copper. . .)” (Zones, 1:61-63.) Thus, when making the catalysts of Zones, one of ordinary skill in the art would be required to consider the amount of copper or other active metal to include.

Ex. 1008, Lercher Decl. at ¶ 152

# Petitioner Asserts That Increasing Cu/Al Ratio Predictably Enhances Zeolite Properties

153. As of February 2007, it was well known in the prior art that copper should be beneficially incorporated into a catalyst in an amount approaching a 100% ion-exchange rate. It was also well known that up to a point, increased amounts of copper, including that achieved by a utilizing a 100% ion-exchange ratio or rate, enhance the effectiveness of a zeolite when catalyzing the reduction of nitrogen oxides. This is stated in Maeshima, which explains that when incorporating copper into a zeolite, “it is generally preferred that the ion exchange ratio be about 60 to about 100%.” (Maeshima, 4:44-54.) Use of these known ion exchange ratios with Zones’ zeolites produces a catalyst with the claimed CHA crystal structure, SAR, and Cu/Al ratio that can be used as part of a process to selectively reduce nitrogen oxides in a gas stream that contains oxygen, nitrogen, and H<sub>2</sub>O.

Ex. 1008, Lercher Decl. at ¶ 153



# Petitioner's Evidence of Predictability

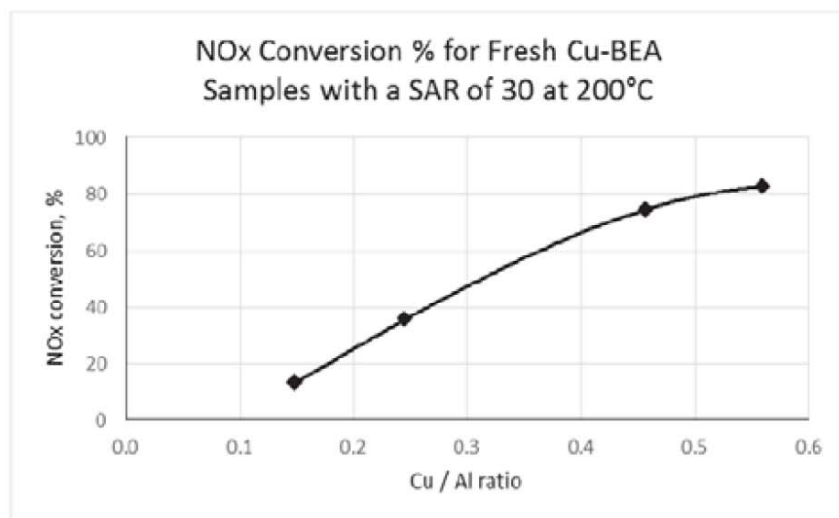


Figure 6

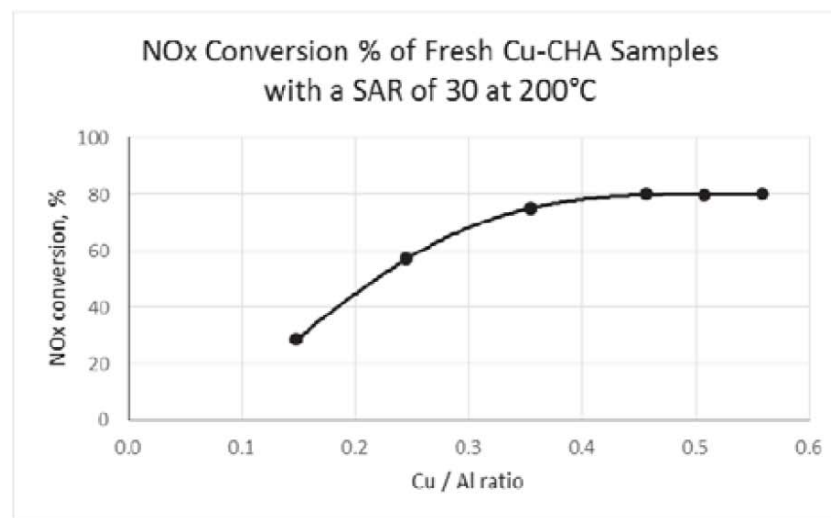
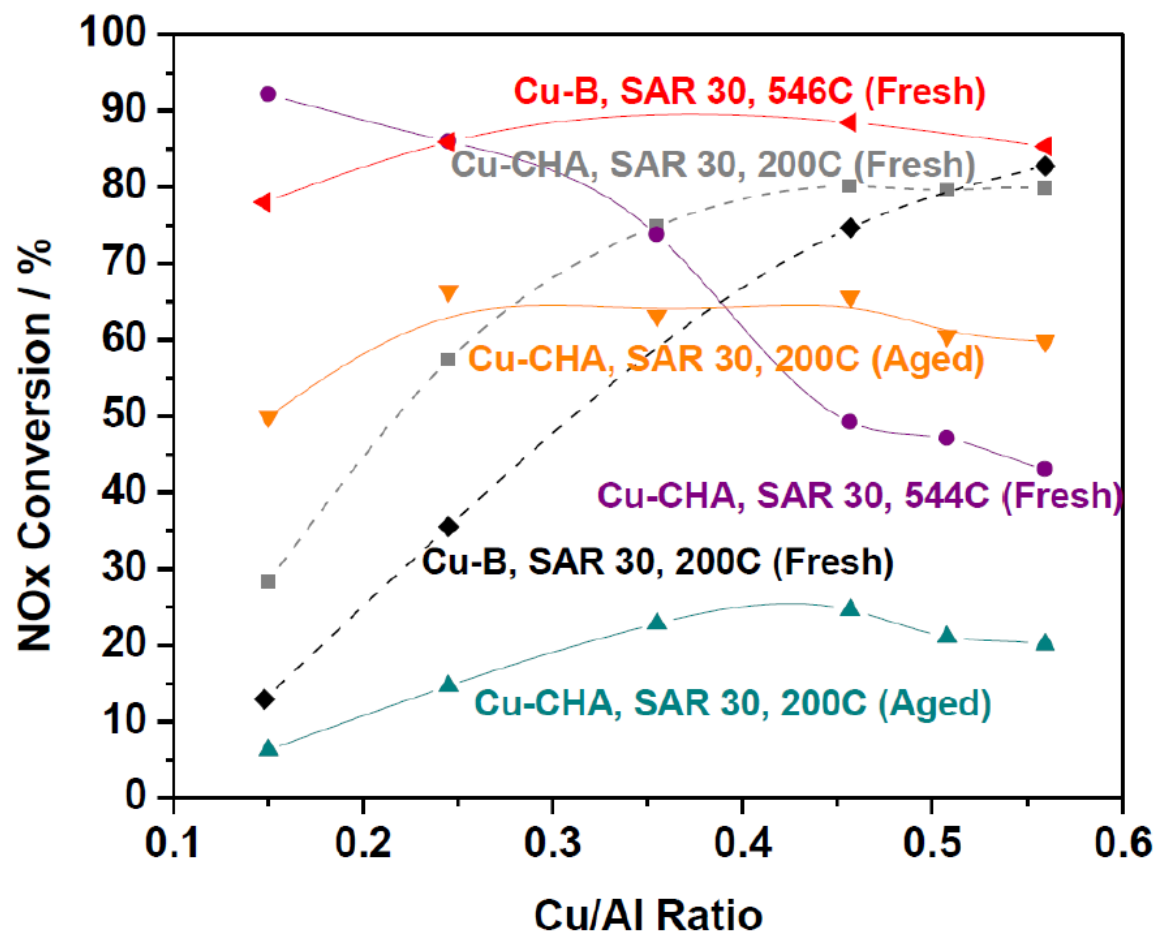


Figure 2

# Unpredictability of Increasing Amount of Copper

**Cu-CHA and Cu-BEA, SAR 30, Cu/Al from 0.1 to 0.6**





# Unpredictability of Increasing Amount of Copper

- Ex. 2012.005 (1995)
  - “A low hydrothermal stability, in particular, is the more critical weakness of copper-containing zeolites.”
- Ex. 2002.001 (2008)
  - “Fe/zeolite formulations are known to exhibit superior hydrothermal stability over Cu/zeolite formulation”
- Ex. 2022.026 (2008)
  - “Iron-exchange ZSM-5 has received much attention because of its promising activity and stability in the  $\text{NH}_3$ -SCR process. The SCR activity of Fe-ZSM-5 has even shown to exceed that of the established commercial  $\text{V}_2\text{O}_5$ - $\text{WO}_3$ - $\text{TiO}_2$  catalysts. Cu-containing zeolites are also very active, though they suffer from low hydrothermal stability and high  $\text{NH}_3$  oxidation activity.”

# Petitioner Correlates The Metal Percentages in Zones and Maeshima

157. As recognized in the '662 patent, “[m]etal-promoted zeolite catalysts, including, among others, iron-promoted and copper-promoted zeolite catalysts, for the selective catalytic reduction of nitrogen oxides with ammonia are known.” ('662 patent, 1:30-34.) Both Zones and Maeshima disclose these types of zeolite catalysts. Zones discloses zeolites with the CHA crystal structure and explains that these zeolites can be ion-exchanged or impregnated with metals like copper. Zones also states that a zeolite may contain 0.05-5% metal cation by weight. Maeshima likewise discloses zeolites with the CHA crystal structure that can be ion-exchanged or impregnated with copper. And, Maeshima states that copper can be incorporated into a zeolite to achieve a 60-100% ion-exchange rate, or such that there is 2-10% active metal by weight. The overlap in these two references’ disclosures would not only motivate one of ordinary skill in the art to combine Zones and Maeshima, but would also lead them to believe that the combination would be successful.

Ex. 1008, Lercher Decl. at ¶ 157

## Zones '644: Metal Weight Unrelated to SCR of NO<sub>x</sub>

### Condensation of Alcohols

SSZ-62 can be used to condense lower aliphatic alcohols having 1 to 10 carbon atoms to a gasoline boiling point hydrocarbon product comprising mixed aliphatic and aromatic hydrocarbon. The process disclosed in U.S. Pat. No. 3,894,107, issued Jul. 8, 1975 to Butter et al., describes the process conditions used in this process, which patent is incorporated totally herein by reference.

The catalyst may be in the hydrogen form or may be base exchanged or impregnated to contain ammonium or a metal cation complement, preferably in the range of from about 0.05 to 5% by weight. The metal cations that may be present include any of the metals of the Groups I through VIII of the Periodic Table. However, in the case of Group IA metals, the cation content should in no case be so large as to effectively inactivate the catalyst, nor should the exchange be such as to eliminate all acidity. There may be other processes involving treatment of oxygenated substrates where a basic catalyst is desired.

Ex.1004 at 1:5-15

# Petitioner Asserts That Zones and Maeshima Are Directed To The Same Problem

158. Additionally, based on my review of Zones and Maeshima, it is my opinion that these two references are also in the same technical field. Both generally relate to zeolite catalysts and the use of these catalysts. Zones and Maeshima are also directed to solving the same problem, catalyzing the reduction of nitrogen oxides. In my opinion, this would further motivate combination of Zones and Maeshima and provides an additional reason why one of ordinary skill in the art would consider it reasonably likely that the combination would succeed.

Ex. 1008, Lercher Decl. at ¶ 158

# Petitioner Asserts That Improving Zeolite SCR Performance is Simple and Straightforward

159. In my opinion, the combination of Zones and Maeshima also amounts to the simple and straight forward application of one particular known modification to catalytic zeolites that provides a known benefit—increasing the amount of copper contained in an aluminosilicate zeolite to improve its catalytic activity—to the exact type of material this modification is meant to be applied—Zones’ high silica, copper-exchanged chabazite zeolites intended for use in an SCR process.

Ex. 1008, Lercher Decl. at ¶ 159

## Zones '538: Metal-Exchanged High SAR, CHA Zeolite

- Metal-exchanged high SAR synthetic CHA zeolite was known in the art **two decades before** Zones '644

“SSZ-13 zeolites can have a  $\text{YO}_2:\text{W}_2\text{O}_3$  mole ratio greater than about 5:1. As prepared, the silica:alumina mole ratio is typically in the range of 8:1 to about 50:1; higher mole ratios can be obtained by varying the relative ratios of reactants.”

Ex.1016 at 2:53-58

“Usually, it is desirable to remove the alkali metal cation by ion exchange and replace it with hydrogen, ammonium, or any desired metal ion.”

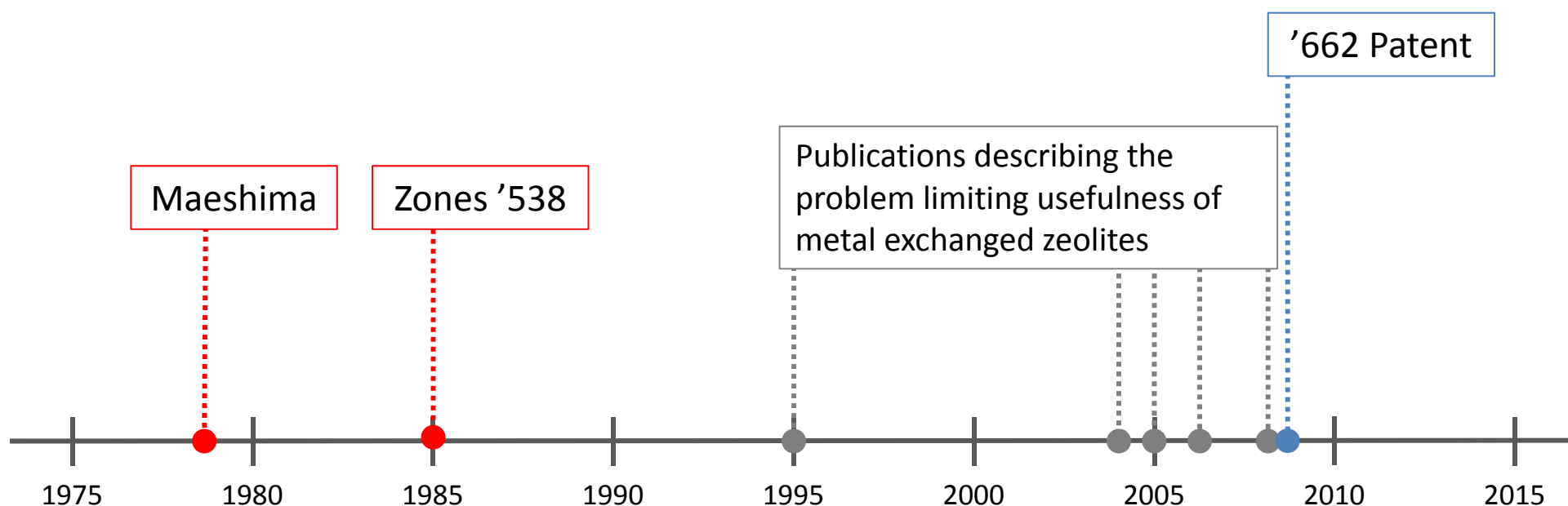
Ex.1016 at 5:41-44



# Timeline Does Not Support Petitioner's Assertion

“The elapsed time between the prior art and the '013 patent's filing date evinces that the '031 patent's claimed invention was not obvious to try. Indeed this considerable time lapse suggests instead that the Board only traverses the obstacles to this inventive enterprise with resort to hindsight.”

*Leo Pharm. Prods. v. Rea*, 726 F.3d 1346, 1356-1357 (Fed. Cir. 2013)



## **Zones + Maeshima + Patchett 843**

('662 Claims 12-24, 32-50)

('203 Claims 2-13, 16, 23-25, 28-31)



# Petitioner Acknowledges The Zeolite Requirements Specified in Patchett '843

246. According to Patchett '843, zeolites that are “resistant to sulfur poisoning” and “sustain a high level of activity for the SCR process” when exposed to “high temperatures” and “hydrothermal conditions” should be used in its system.  
(Patchett '843, ¶ 66.)

Ex. 1008, Lercher Decl. at ¶ 246

## Petitioner Asserts That Patchett '843 Would Direct POSITA to CHA Zeolite

249. I have reviewed these references, which include U.S. 4,961,917 to Byrne and U.S. 5,516,497 to Speronello et al. They provide examples of zeolites with SARs greater than 10. (*See, e.g.*, U.S. 4,961,917 to Byrne at Abstract, 2:26-35; U.S. 5,516,497 to Speronello et al at Abstract, 6:3-7:3.)

250. In my opinion, in view of this discussion of zeolite materials in Patchett '843, one of ordinary skill in the art as of February 2007 would be directed to the materials of Zones and Maeshima, and would consider those materials for use in Patchett '843's system.

Ex. 1008, Lercher Decl. at ¶¶ 249-250

# Patchett '843 Teaches Away from CHA Zeolites

nected in three dimensions. Without wishing to be bound by any specific theory, it is believed that the interconnection of pores of at least 7 Angstroms diameter in three dimensions provides for good mobility of sulfate molecules throughout the zeolite structure, thereby permitting the sulfate molecules to be released from the catalyst to free a large number of the available adsorbent sites for reactant NO<sub>x</sub> and NH<sub>3</sub> molecules and reactant NH<sub>3</sub> and O<sub>2</sub> molecules. Any zeolites meeting the foregoing criteria are suitable for use in the practices of the present invention; specific zeolites which meet these criteria are USY, Beta and ZSM-20. Other

Ex.1005 at [0066]

# Patchett '843 Teaches Away from CHA Zeolites

## SCR Catalyst Compositions

[0065] Suitable SCR catalyst compositions that may be used to coat the inlet zone of the first substrate and/or the second substrate (in embodiments depicted in **FIG. 3B**) are described, for instance, in **U.S. Pat. No. 4,961,917** (the '917 patent) and **U.S. Pat. No. 5,516,497** (the '497 patent), which are both hereby incorporated by reference in their entirety.

Ex.1005 at [0065]

# Patchett '843 Teaches Away from CHA Zeolites

- Speronello (Ex. 1011 at 6:43-59)
  - “Specifically, an average pore size of about 7 Angstroms or more, e.g., about 7 to about 8 Angstroms is preferred for enhanced resistance to sulfur poisoning...[A] particularly suitable class of such sulfur-resistant zeolite materials is comprised of Beta zeolites, ultrastable Y (“USY”) zeolites, and ZSM-20 zeolites.”

# Patchett '843 Teaches Away from CHA Zeolites

- Byrne (Ex. 1010 at 4:57-5:26, 6:12-32)
  - “For example, flowing a gas stream containing 2,000 parts per million by volume ("Vppm") SO<sub>2</sub> through catalysts comprising copper-promoted small to medium pore zeolites such as ZSM-5, naturally occurring chabazite and clinoptilolite, resulted in 10 to 40 percent reduction in SCR process activity. Even at SO<sub>2</sub> levels as low as 130 Vppm SO<sub>2</sub>, significant activity reduction for the SCR process was noted for such catalysts....In the case of the small to medium pore zeolites, this competition absorption with NH<sub>3</sub> and NO probably results in a physical blockage and/or diffusional restriction....A 60 percent reduction in SCR process activity is typical for Fe<sub>2</sub> O<sub>3</sub> containing natural chabazite.”
  - “It has been found that zeolites which are highly resistant to sulfate poisoning and provide good activity for both the SCR process ....are zeolites which have pores which exhibit a pore diameter of at least about 7 Angstroms and are interconnected in three dimensions....[S]pecific zeolites which meet these criteria are USY, Beta, and ZSM-20.”

## Petitioner Disregards Relevant Parts of Prior Art

“This type of reasoning—where relevant parts of the reference are disregarded for the proposed combination without sufficient explanation of why a person of ordinary skill would do so—is precisely the type of hindsight reasoning that must be rejected.”

*Oracle Corp., et al., v. Crossroads Systems, Inc.*, IPR2014-01207, Paper 78 at 37 (Jan. 29, 2016)

# Patchett '514: Wall Flow Filter

- '662 claims 13, 18-20, 23, 24, 39, 44-46, 49, 50
- '203 claims 5-7, 24, 30

[0011] An additional aspect for consideration in coating the wall flow filter is the selection of the appropriate SCR catalyst composition. First, the catalyst composition must be durable so that it maintains its SCR catalytic activity even after prolonged exposure to higher temperatures that are characteristic of filter regeneration. For example, combustion of the soot fraction of the particulate matter often leads to temperatures above 700° C. Such temperatures render many commonly used SCR catalyst compositions such as mixed oxides of vanadium and titanium less catalytically effective. Second, the SCR catalyst compositions preferably have a wide enough operating temperature range so that they can accommodate the variable temperature ranges over which the vehicle operates. Temperatures below 300° C. are typically encountered, for example, at conditions of low load, or at startup. The SCR catalyst compositions are preferably capable of catalyzing the reduction of the NO<sub>x</sub> component of the exhaust to achieve NO<sub>x</sub> reduction goals, even at lower exhaust temperatures.

Ex.1006 at [0011]



# Zones '644 and Maeshima: No Disclosure of Stability at High Temperatures

## Maeshima

“As the reaction conditions, there may be adopted a reaction temperature of about 200° to about 500° C, preferably about 250° to about 400° C, and a gas space velocity of about 2,000 to about 100,000 V/H/V, preferably about 5,000 to about 30,000 V/H/V. Since the activity of the ammonia reduction of nitrogen oxides is lowered at higher or lower temperatures, good results are obtained when a mixture of the exhaust gas and ammonia is contacted with the catalyst bed at a temperature within the above-mentioned range.”

Ex.1004 at 7:16-23

## Zones '644



Ex.1004

# Zones '644 and Maeshima: No Disclosure of Stability at High Temperatures

## Maeshima

As the reaction conditions, there may be adopted a reaction temperature of about 200° to about 500° C., preferably about 250° to about 400° C... Since the activity of the ammonia reduction of nitrogen oxides is lowered at higher or lower temperatures, good results are obtained when a mixture of the exhaust gas and ammonia is contacted with the catalyst bed at a temperature within the above-mentioned range.

Ex. 1002 at 3:23-32

## Petitioner's Expert on Zones

Q: A slightly broader question: Does the Zones patent specifically recognize the problem of hydrothermal stability of a zeolite for the SCR of NOX?

[Form Objection]

A: I do understand your question, but you're asking whether Zones is referring to the problem of hydrothermal stability with respect to NOX removal.

Q: Yes.

A: I do not see this in this document.

Ex. 2027 at 48:5-17



## **Maeshima + Breck**

('662 Claims 1, 2, 5, 6, 30)

('203 Claims 1, 14, 15, 19, 20, 26, 27)

# Breck: Dealumination of Zeolites

93. Breck teaches a method to increase the SAR of a zeolite through dealumination. De-alumination is a method of increasing the SAR by removing aluminum from the zeolite framework and replacing the aluminum with silicon from an extraneous source. Exhibit 1003 at 3:24-31. The ending SAR of a zeolite that is subjected to the de-alumination process is at least a function of the starting SAR and the zeolite framework type, which impacts the efficiency at which aluminum ions can be removed. The specific examples that Breck teaches, and the related discussion, show these known limitations, which Dr. Lercher does not address in his declaration.

Ex. 2018 at ¶93

# Breck: Dealuminating Zeolites

Example No.	Zeolite	Starting SAR	Ending SAR
1	Y	4.85	9.54
2	Y	4.8	9.31
3	Y	4.97	11.98
5A	Y	3.50	4.84
5B	X	2.52	6.43
6	A	5.43	7.38
9A	L	6.03	12.39
9B	L	5.92	11.73
11	Chabazite	8.52	11.13
12	Erionite	6.96	9.77
14	W	3.88	11.67
15	Rho	5.92	12.36
16	Y	4.93	14.80
17	Y	4.93	11.72
18	Y	4.93	7.30
19	Omega	6.20	6.67
20	Y	4.87	9.87

Ex. 2018 at ¶96

## Breck: Aluminum Removal Inefficient for Chabazite

The data indicate that silicon has replaced aluminum in the chabazite framework. The efficiency of aluminum removal is relatively low compared with the Y, L and mordenite zeolites, and comparable to that observed in the case of clinoptilolite. However with chabazite, sili-

Ex.1003 at 38:44-48

# Maeshima: Zeolite Frameworks

Suitable natural zeolites are:

Mordenite:  $(\text{Ca}, \text{K}_2, \text{Na}_2) [\text{AlSi}_5\text{O}_{12}]_2 \cdot 7\text{H}_2\text{O}$

Erionite:  $(\text{K}_2, \text{Na}_2, \text{Ca}) [\text{AlSi}_3\text{O}_8]_2 \cdot 6\text{H}_2\text{O}$

Natrolite:  $\text{Na}_2[\text{Al}_2\text{Si}_3\text{O}_{10}] \cdot 2\text{H}_2\text{O}$

Chabazite:  $(\text{Ca}, \text{Na}_2) [\text{Al}_2\text{Si}_4\text{O}_{12}] \cdot 6\text{H}_2\text{O}$

Faujasite:  $\text{Na}_2\text{Ca} [\text{Al}_2\text{Si}_4\text{O}_{12}]_2 \cdot 16\text{H}_2\text{O}$

The natural zeolites contain alkaline earth metals, with alkali metals.

As the synthetic zeolites, there may be used synthetic faujasite and synthetic mordenite. The synthetic faujasites include:

Zeolite-A:  $1.0 \pm 0.2\text{M}_{2/n}\text{O} : \text{Al}_2\text{O}_3 : 1.85 \pm 0.5\text{SiO}_2 : \text{YH}_2\text{O}$  (wherein M represents a metal,  $n$  represents a valence of M and Y represents a number of about 6 or below).

Zeolite-X:  $1.0 \pm 2\text{M}_{2/n}\text{O} : \text{Al}_2\text{O}_3 : 5 \pm 0.5\text{SiO}_2 : \text{YH}_2\text{O}$  (wherein M represents a metal of a valence of from 1 to 3, inclusive,  $n$  represents a valence of M and Y represents a number of about 8 or below).

Zeolite-Y:  $0.9 \pm 0.2\text{Na}_2\text{O} : \text{Al}_2\text{O}_3 : \text{WSiO}_2 : \text{YH}_2\text{O}$  (wherein W represents a number between 3 and 6, inclusive, and Y represents a number of about 9 or below).

The synthetic mordenites include, for example:

Zeolite-L:  $1.0 \pm 0.1\text{M}_{2/n}\text{O} : \text{Al}_2\text{O}_3 : 0.4 \pm 0.5\text{SiO}_2 : \text{YH}_2\text{O}$  (wherein M represents a metal,  $n$  represents a valence of M, and Y represents a number from 0 to 7, inclusive).

Ex.1002 at 4:6-35

# Maeshima: Use Low SAR, Large Pore Size Zeolite Frameworks

Especially preferred crystalline aluminosilicates are those having a pore diameter in the range of about 6–13 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 2–6. For example, synthetic faujasite having a pore diameter of about 8–9 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 2–3 and other synthetic faujasites having a pore diameter of about 8–9 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 4–6 are preferred.

Ex.1002 at 4:36-43



# Petitioner Asserts That Dealumination Will Not Detrimentally Impact Zeolite Activity

166. In my opinion, one of ordinary skill in the art would also have a reasonable expectation of success when combining the teachings of Maeshima and Breck.

167. Increasing the SAR of the zeolite utilized by Maeshima would not detrimentally impact the usefulness of that zeolite in a process for reducing nitrogen oxides.

Ex. 1108, Lercher Decl. at ¶¶ 166-167

# Dr. Lercher Cross-Examination

Q: Does de-aluminating the zeolite impact the activity or stability of the zeolite?

A: It impacts both. As we have said before, the concentration of aluminum in the lattice determines how many ion exchange positions you have, or in the case of an acid material, how many protons you have. Therefore, the rates that you have are in the first approximation directly proportional to that concentration. In opposite, for gas phase reactions, the concentration of aluminum has an adverse effect on the hydrothermal stability of that zeolite in two ways: One is you remove a particular aluminum out of that lattice. That means when you treat it by steam, you just have less elements. On the other side, if you have -- like, you should see that lattice like a building. If you remove too many bricks, your whole building collapses, so structures of that zeolite collapse.

Q: So, in a sense, de-illuminating the zeolite can decrease the activity but may improve the stability?

A: Yes.

Ex. 2027 at 94:23-95:25

# Dr. Tsapatsis Declaration

122. Furthermore, a person of ordinary skill would not have a reasonable expectation that de-aluminating a zeolite to increase the SAR would result in an active and stable zeolite for the SCR of NO<sub>x</sub>. Although Breck does contend that higher SAR may improve stability, there is a diverging effect of de-alumination that is not addressed by Breck. Specifically, the removal of aluminum leaves fewer extra framework ions present in the catalyst, thereby reducing the catalytic activity of the zeolite. Thus, while de-alumination may improve stability, it can also destroy the catalytic activity of the zeolite. Dr. Lercher did not address this issue in his declaration, but he acknowledged the opposing effects of de-alumination at his deposition. Exhibit 2027 at 93:8-95:25.

Ex. 2018 at ¶122

# Dr. Tsapatsis Declaration

123. In addition, the 662 Patent itself states the decline in the catalytic activity of a zeolite after hydrothermal aging appears to be due to de-alumination: “This decline in activity is believed to be due to destabilization of the zeolite such as by dealumination and consequent reduction of metal-containing catalytic sites within the zeolite.” 662 Patent at 1:38-41. Accordingly, it cannot be said that the impact of de-alumination is predictable with respect to the activity and stability of a zeolite catalyst.

Ex. 2018 at ¶123

# Maeshima: Starting SAR

Especially preferred crystalline aluminosilicates are those having a pore diameter in the range of about 6–13 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 2–6. For example, synthetic faujasite having a pore diameter of about 8–9 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 2–3 and other synthetic faujasites having a pore diameter of about 8–9 Å and  $\text{SiO}_2/\text{Al}_2\text{O}_3$  molar ratio of about 4–6 are preferred.

Ex.1002 at 4:36-43



## Breck: Increasing SAR of Chabazite is Not Efficient

	Starting NH <sub>4</sub> — Chabazite	LZ-218 Product
Al <sub>2</sub> O <sub>3</sub> , wt. %	14.83	12.05
SiO <sub>2</sub> , wt. %	74.51	78.98
F <sub>2</sub> , wt. %	NA	0.39
SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub>	8.52	11.13
Na <sup>+</sup> /Al	NA	0.12
NH <sub>4</sub> <sup>+</sup> /Al	0.66	0.54
Cation Equivalent, M <sup>+</sup> /Al	0.66	0.94

Ex.1003 at 38:1-10

The data indicate that silicon has replaced aluminum in the chabazite framework. The efficiency of aluminum removal is relatively low compared with the Y, L and mordenite zeolites, and comparable to that observed in the case of clinoptilolite. However with chabazite, sili-

Ex.1003 at 38:44-48

# Petitioner Asserts That Breck and Maeshima Are Directed To The Same Problem

169. Additionally, based on my review of Maeshima and Breck, it is my opinion that these two references are also in the same technical field. Both generally relate to zeolite catalysts and the use of these catalysts. Breck and Maeshima are also directed to solving the same problem, identifying materials that can be effectively used in a process that, for instance, catalyzes the reduction of nitrogen oxides. In my opinion, this would further motivate combination of Breck and Maeshima and provides an additional reason why one of ordinary skill in the art would consider it reasonably likely that the combination would succeed.

Ex. 1108, Lercher Decl. at ¶ 169

## Petitioner Asserts That Increasing SAR Predictability Increases SCR Activity

411. It was also well known as of February 2007 that increasing the SAR of a zeolite renders it more hydrothermally stable and more resistant to the effects of hydrothermal aging.

Ex. 1108, Lercher Decl. at ¶¶ 411-421



# Prior Art: Increasing SAR Can Decrease Activity

## SELECTIVE CATALYTIC REDUCTION OF $\text{NO}_x$ OVER ACID-LEACHED MORDENITE CATALYSTS

Lars A.H. Andersson, Jan G.M. Brandin and C.U. Ingemar Odenbrand  
Department of Chemical Technology, Chemical Center, Lund University,  
Institute of Science and Technology, P.O. Box 124, S-221 00 Lund, (Sweden)

Ex.2025.001

## CONCLUSIONS

### Conclusions on $\text{NO}_x$ reductions over acid leached mordenite

1. The catalytic activity of mordenite decreases with increasing Si/Al atomic ratio in the reductions of NO and of  $\text{NO}_x$  at  $\text{NO}_2/\text{NO}_x = 0.45$ .

Ex.2025.012

## Prior Art: Increasing SAR Can Decrease Activity

The analysis of NO reduction by  $\text{NH}_3$  in the presence of  $\text{O}_2$  shows a continuous NO reduction. The Si/Al ratio determines the number of Brønsted acid sites in ZSM-5 and therefore also the capacity to introduce copper ions at exchangeable sites [18]. The Cu-ZSM-5-27 catalyst holds the largest number of acid sites and the result shows that this catalyst, with the lowest ratio, reveals the highest  $\text{NO}_x$  conversion, and, as the ratio increases, the conversion decreases. Similar Si/Al effects were shown by Long and Yang [19], who reported decreasing NO conversions with increasing Si/Al ratio over Fe-ZSM5 catalysts with similar ion-exchange levels. Komatsu et al. [11] reported that a decreased Si/Al ratio improves the specific activity (per  $\text{Cu}^{2+}$  ion) over Cu-ZSM-5, because the aluminum concentration increases resulting in an increase in the copper concentration.

Ex.2023.004

# Petitioner Asserts That Improving Zeolite SCR Performance is Simple and Straightforward

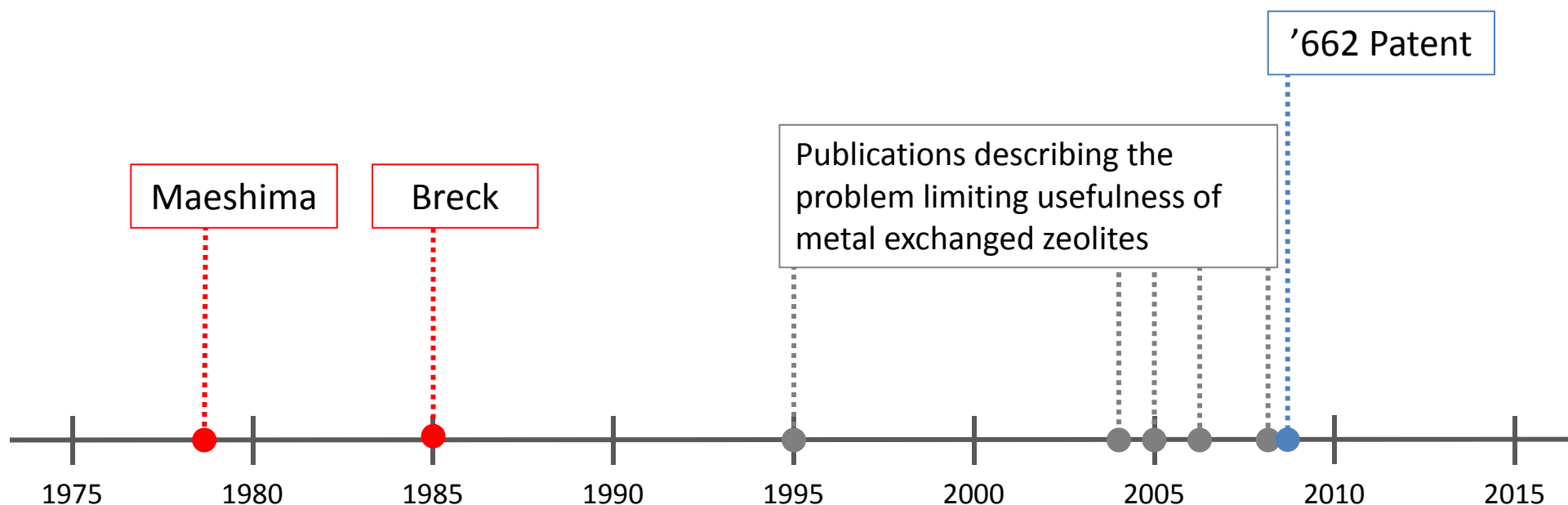
170. In my opinion, the combination of Maeshima and Breck also amounts to the simple and straight forward application of one particular known type of catalytic material with known benefits, the higher SAR chabazite zeolites of Breck, to a process that calls for the use of just this material, Maeshima's process for selectively reducing nitrogen oxides.

Ex. 1108, Lercher Decl. at ¶ 170

# Timeline Does Not Support Petitioner's Assertion

“The elapsed time between the prior art and the '013 patent's filing date evinces that the '031 patent's claimed invention was not obvious to try. Indeed this considerable time lapse suggests instead that the Board only traverses the obstacles to this inventive enterprise with resort to hindsight.”

*Leo Pharm. Prods. v. Rea*, 726 F.3d 1346, 1356-1357 (Fed. Cir. 2013)





## **Dedecek + Breck**

('662 Claims 1, 2, 5, 6, 30)

('203 Claims 1, 14, 15, 19, 20, 26, 27)

## Dedecek: Study About Cu<sup>+</sup> Ion Siting

- **Title:** *Siting of the Cu<sup>+</sup> ions in dehydrated ion exchanged and natural chabasites: a Cu<sup>+</sup> photoluminescence study*

[14,15]. But there is still a lack of general information on the siting and coordination of the Cu<sup>+</sup> ions in zeolites.

Ex.1007.001

# Dedecek: ZSM-5 for SCR of NO<sub>x</sub>

“Zeolites containing Cu ions attract attention owing to their high catalytic activity in NO [1-5] and N<sub>2</sub>O decomposition [6] and selective catalytic reduction (SCR) of NO with ammonia [7-9] and hydrocarbons [10-12].”

Ex.1007.001

[7] B. Wichterlová, Z. Sobalí, A. Vondrová, Catal. Today 29 (1996) 149.

[8] T. Komatsu, M. Nunokawa, I.S. Moon, T. Takahara, S. Namba, Y. Yashima, J. Catal. 148 (1994) 427.

[9] J.A.S. Sullivan, J. Cunningham, M.A. Morris, K. Ken-  
eavey, Appl. Catal. B: 7 (1995) 137.

MFI framework  
[ZSM-5]

Ex.2028, 2029, 2030

# Petitioner's Expert Testimony Regarding Dedeczek

Q: Do you agree that there is no discussion, teaching or data in Dedeczek regarding the use of copper chabazite in any one of those reactions?

[Form Objection]

A: Dedeczek describes materials; he does not address reactions.

Q: Okay.

A: He links to those reactions through his introduction.

Q: That's the only place --

A: That's the only place.

Q: -- in the first sentence of page 1?

A: Yes.

Ex.2027 at 79:12-25



# Petitioner Asserts That Dealumination Will Not Detrimentially Impact Zeolite Activity

345. Increasing the SAR of the zeolite utilized by Dedecek would not detrimentally impact the usefulness of that zeolite in a process for reducing nitrogen oxides.

Ex. 1108, Lercher Decl. at ¶ 345

# Dedecek: Starting SAR

276. Dedecek references on “batch composition” of a “chabasite” that “was 0.17Na<sub>2</sub>O:2.0K<sub>2</sub>O:Al<sub>2</sub>O<sub>3</sub>:5.4SiO<sub>2</sub>:224H<sub>2</sub>O” (Dedecek, p. 64.) This material has a SAR of 5.4. Dedecek also references “chemical composition (weight percent)” of a chabazite material that was “63.89% SiO<sub>2</sub>, 17.48% Al<sub>2</sub>O<sub>3</sub>, 8.37% Fe<sub>2</sub>O<sub>3</sub>, 5.15% K<sub>2</sub>O, 3.10% CaO, 1.21% MgO, 0.40 TiO<sub>2</sub> and 0.39% Na<sub>2</sub>O.” (Dedecek, p. 64.) Since this material is a chabazite, it will have the CHA crystal structure. Further, the SAR of the zeolite can be calculated using the provided weight percents:

$$63.89 \text{ g SiO}_2 \div 60.08 \text{ g/mol SiO}_2 = 1.06 \text{ mol SiO}_2$$

$$17.48 \text{ g Al}_2\text{O}_3 \div 101.96 \text{ g/mol Al}_2\text{O}_3 = 0.17 \text{ mol Al}_2\text{O}_3$$

$$1.06 \text{ mol SiO}_2 \div 0.17 \text{ mol Al}_2\text{O}_3 = \text{SAR } 6.2$$

Ex. 1108 at ¶276

# Petitioner Asserts That Dedecek Contemplates Higher SAR

339. Dedecek itself would further motivate one of ordinary skill in the art to consider the use of higher SAR zeolites, including those set forth in Breck. While Dedecek provides examples of chabazite zeolites with SARs of 5.4 and 6.2, these are just examples. Dedecek provides no upper bound on the useful SAR of a zeolite. In fact, Dedecek appears to just be referencing the specific materials that were purchasable at the time the authors engaged in the testing reported in Dedecek. Thus, the disclosure of Dedecek is open to and contemplates that higher SAR zeolites can be used.

Ex. 1108, Lercher Decl. at ¶ 339

# Petitioner Asserts That Breck and Maeshima Are Directed To The Same Problem

347. Additionally, based on my review of Dedecek and Breck, it is my opinion that these two references are also in the same technical field. Both generally relate to zeolite catalysts and the use of these catalysts. Dedecek and Breck are also directed to solving the same problem, identifying materials that can be effectively used in a process that, for instance, catalyzes the reduction of nitrogen oxides. In my opinion, this would further motivate combination of Dedecek and Breck and provides an additional reason why one of ordinary skill in the art would consider it reasonably likely that the combination would succeed.

Ex. 1108, Lercher Decl. at ¶ 347



## **Maeshima/Dedecek + Breck + Patchett '843**

('662 Claims 12-24, 32-50)

('203 Claims 2-13, 16, 23-25, 28-31)

## **Secondary Considerations**

# Commercial Success: Product Specification

- **BASF's CuCHA Product (Ex. 2019)**
  - SAR: 28-34 (target 31)
  - Cu/Al: 0.38-0.42
  - NH<sub>3</sub>-SCR of NO<sub>x</sub> in diesel engine
  - Sold on flow-through substrate or wall-flow filter
- **Claims Practiced (Ex. 2018 at ¶ 175-177)**
  - 662 Claims: 1-8, 12-15, 18, 21, 23, 39, 40, 41, 44, 47, 50
  - 203 Claims: 1-3, 5, 6, 14-24, 26-30
- **Market Size and Share**
  - Ex. 2034 (Schmidt Declaration)

## Commercial Success: Market Size and Share

- **Estimated Global Diesel SCR Market (Ex. 2034):**

Total Global SCR Market by Units Sold (in million)		
2011		■
2012		■
2013		■
2014		■
2015		■

- **CuCHA Estimate Market Share (Ex. 2034)**

- ■ (combined BASF and 662 licensee)



## **CERTIFICATE OF SERVICE**

The undersigned hereby certifies that on July 25, 2016 the foregoing  
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2015-01124 AND 2015-01125 was served via electronic mail, upon the following:

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