IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Inter Partes Reexamination of:)	••
	٥	Examiner: DIAMOND, ALAN D
BULL ET AL.)	
		Group Art Unit: 3991
Reexamination Control No. 95/001,453)	•
	1	Confirmation No: 2755
Patent No. 7,601,662)	
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Issued: October 13, 2009)	
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For: COPPER CHA)	
ZEOLITE CATALYSTS	*	

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Central Reexamination Unit Commissioner for Patents P.O. Box 1450 Alexandria, VA 22313-1450

DECLARATION OF STACEY I. ZONES, PH.D. UNDER 37 C.F.R. § 1.132

I, Stacey Zones, do declare and say as follows:

- 1. I have a Ph.D. in Chemistry from the University of California at San Diego. I am a Research Fellow at Chevron Energy Technology Company, a division of Chevron U.S.A. Inc. a wholly owned subsidiary of Chevron Corporation (ETC). I have been involved in research and development of molecular sieves and zeolites at ETC for more than thirty years. In 2001, I received the Donald W. Breck award from the International Zeolite Association for my work related to molecular sieves. In 2007, I received the Eugene J. Houdry award in Applied Catalysis from the North American Catalysis society for my work on zeolite catalysts. I am named as an inventor on 115 United States granted patents pertaining to the synthesis and/or use of molecular sieves. I am the first-named inventor on United States patent number 6,709,644 ("the '644 patent").
- 2. I am informed that United States patent number 7,601,662, ("the '662 patent"), assigned on its face to BASF Catalysts LLC ("BASF"), is currently under reexamination in the United States Patent and Trademark Office. I am also informed that the '644 patent is cited as a reference in rejecting the claims of the '662 patent in the above captioned reexamination of the '662 patent.



- 3. Representatives of BASF Corporation, the owner of the '662 patent, contacted me and requested that I provide the statements and expert opinion contained in this Declaration.
- 4. I am aware and understand that my employer, Chevron U.S.A. Inc., and BASF Corporation are in several business relationships related to the sale and manufacture of catalysts, including a license to the '644 patent.
- 5. I have not requested and I'm not receiving any special or additional compensation from my employer in connection with this Declaration and I have not requested and I am not being compensated by the owner of the '662 patent or any other person for the preparation of this Declaration.
- 6. BASF Corporation representatives asked me to provide my understanding and opinion regarding the teachings of the '644 patent. The '644 patent pertains to a zeolite having the crystal structure of chabazite (CHA) having small crystallite size, processes using the small crystallite CHA as a catalyst and gas separation processes using the small crystallite CHA.
- 7. The '644 patent discloses that a zeolite having the CHA crystal structure can be used for a variety of processes including separation of gasses, such as separating carbon dioxide from natural gas (col. 5, lines 66-67), as catalysts used for the reduction of oxides of nitrogen in a gas stream in the presence of oxygen (col. 1, lines 54-66), converting lower alcohols and other oxygenated hydrocarbons to a gasoline boiling point hydrocarbon product (col. 5, lines 18-14), and for producing dimethylamine (col. 5, lines 36-40). Additionally, the '644 patent states that a zeolite having the CHA crystal structure may contain a metal or metal ions (such as cobalt, copper or mixtures thereof) capable of catalyzing the reduction of oxides of nitrogen, and may be conducted in the presence of a stoichiometric excess of oxygen. (col. 1, lines 61-65).
- 8. In my view, the phrase "reduction of oxides of nitrogen contained in a gas stream in the presence of oxygen" in the '644 patent refers to and teaches a number of different reactions, including the reduction of NO without reducing agents, such as in the presence of CO (for example, a three-way catalyst), the decomposition of NO in the presence of oxygen, and N_2O decomposition, the selective catalytic reduction of NO in the presence of an ammonia reducing agent and oxygen, and the selective catalytic reduction of NO in the presence of a hydrocarbon reducing agent and oxygen. The '644 patent does not teach specific reactions for the reduction of oxides of nitrogen in the presence of oxygen.
- 9. It is generally known in the art that zeolites that contain metal or metal ions, such as copper, cobalt, other metal cations and mixtures thereof, are generally suitable for one or more of these reactions of reducing of oxides of nitrogen in a gas stream in the presence of oxygen. Several patents assigned to Chevron U.S.A. Inc. pertaining to molecular sieves contain statements that the molecular sieve in the patent is useful for the reduction of oxides of nitrogen in the presence of excess oxygen, and that the molecular sieve can contain metal ions such as copper, cobalt, mixtures thereof and other metals. For example, United States patent Nos. 7,138,099 (SSZ-73, SAS framework type); 6,676,732 (SSZ-50, RTH Framework Type); and 6,540,903 (SSZ-47, New Framework Type) are just a few examples of such patents.



- 10. The '644 patent does not specifically disclose or suggest that the zeolite having the CHA crystal structure is useful for selective catalytic reduction of oxides of nitrogen in the presence of a reductant, such as ammonia. There is no discussion, suggestion or example in the '644 patent indicating that a zeolite having the CHA crystal structure containing copper exhibits improved selective catalytic reduction of oxides of nitrogen at temperatures below 350° C compared to other zeolites, or that a zeolite having the CHA crystal structure and containing copper maintains excellent conversion of oxides of nitrogen after hydrothermal aging at temperatures in excess of 650° C and 10% H₂O. I understand that many zeolites containing copper and other metals have had limited application as selective catalytic reduction catalysts in diesel engines because of problems related to maintenance of good nitrogen oxide conversion upon hydrothermal exposure at temperatures in excess of 650° C.
- 11. The '644 patent does not describe or suggest an amount of copper to be used in the zeolite having the CHA crystal structure. Column 5, lines 25-28 of the '644 patent states that the catalyst may contain an ammonium or metal cation complement, preferably in the range from about 0.05% to 5% by weight. This statement appears under the heading of "Condensation of Lower Alcohols" at col. 5, line 17, and the discussion under this heading pertains to a catalyst for the condensation of lower alcohols. In this regard, the '644 patent actually states at column 5, lines 25-33:

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.

This passage quoted above teaches that a catalyst for the condensation of alcohols can include 0.05 to 5% by weight can include metals from Groups I to VIII of the periodic table, which includes all metals in the Periodic Table. The quoted passage of the '644 patent makes reference to group IA metals specifically, but Group IA metals do not include copper. This passage is not related to the passage earlier in the '644 patent at col. 1, lines 61-65 pertaining to reduction of oxides of nitrogen, and there is no teaching in the '644 patent of the amount of copper, cobalt or mixtures thereof that can be used for a catalyst for the reduction of oxides of nitrogen.

12. The '644 patent teaches a small crystal zeolite with the CHA crystal structure having a crystallite size of $0.5~\mu$ or less. Additionally, the '644 patent teaches a method for making the small crystal zeolite, teaches that it is useful for converting lower alcohols and oxygenated hydrocarbons into liquid products and teaches that it is useful for the reduction of oxides of nitrogen in a gas stream in the presence of oxygen, as examples amongst a variety of potential applications of this new material. The' 644 patent does not teach a person having ordinary skill in the art a specific zeolite having the CHA crystal structure, a silica to alumina mole ratio



greater than 15 and a Cu to Al atomic ratio exceeding 0.25 that would be any better than any other zeolite for the reduction of nitrogen oxides in an exhaust gas stream of an internal combustion engine at temperatures below 350° C. The '644 patent does not teach a specific zeolite having the CHA crystal structure that has especially good hydrothermal stability in retaining NOx conversion performance after aging under hydrothermal conditions.

13. I hereby declare that all statements made herein of my own knowledge are true and that all statements made herein on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the patent at issue in this proceeding, US patent number 7,6 01,662.

Dated: February 7, 2011

By:

Stacey I. Zones, Ph.D

