Petitioner

Patent No. 8,404,203
Issue Date: March 16, 2013
Title: PROCESS FOR REDUCING NITROGEN OXIDES USING COPPER CHA ZEOLITE CATALYSTS

DECLARATION OF Dr. FRANK-WALTER SCHÜTZE

Case No. IPR2015-01124

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connection with automotive catalysts at Umicore AG & Co. KG ("Umicore"), which is located in Hanau-Wolfgang, Germany.

2. I studied chemistry at the University of Leipzig (Germany) and received my Ph.D. in Chemistry in 1997. From 1997 to 2001, I was a post doc researcher at the University of Oldenburg (Germany) and the Institute of Applied Catalysis Berlin (Germany).

3. I have held my current position at Umicore since the 1st of January 2015. Prior to that, I was Senior Manager R&D / Research and Customer Projects and was involved in SCR / ASC development. Since I joined Umicore in 2001, I was involved in R&D for automotive catalysts on several topics, very often related to application of zeolites in catalyst formulations.

II. ASSIGNMENT

4. I was asked to make samples of copper-loaded chabazite zeolite ("Cu-CHA") catalysts with varying silica to alumina molar ratios (which I will refer to as the "SAR") and copper to aluminum atomic ratios (which I will refer to as the "Cu/Al ratio").

5. I was asked to test the catalyst samples I made in different ways. In

framework SARs. I obtained chabazite materials with SARs of 13, 19, 21, 27, and 30.

7. Next, I copper-loaded these various NH_4-CHA materials to produce Cu-CHA zeolite samples with different Cu/Al ratios ranging from 0 to 1. Copper-loading of the NH_4-CHA materials was performed by aqueous ion-exchange. The required amount of copper-acetate needed to produce a given Cu/Al ratio was mixed with the NH_4-CHA and the suspension was then heated for 2 hours at 65 °C.

8. For the creation of recipes related to the targeted Cu/Al ratios for CHA materials with the different SAR, I have used molar relationships of the components based on their direct structural correlations. Based on the SAR of the CHA material, I determined the molar composition of the so called "unit cell" (or "u.c.") of the material. The unit cell of a protonated CHA material has the formula ($[Si_{36-x}\ Al_x\ O_{72}]\ H_x$). Using this formula, the molar amount of aluminum or alumina in this structural building unit, and thus the ion-exchange capacity, can be calculated. I used the well accepted stoichiometric assumption that 1 Cu^{2+} ion balance the charge introduced by 2 Al atoms in the structural building unit.

9. For example: a CHA zeolite with an SAR of 22 contains 3 moles of Al in

that can be ion-exchanged into a zeolite with a SAR of 22 is 1.5 mole of Cu, or 95.319 g Cu/mole CHA unit cell.

10. The unit cell formula for a complete stoichiometric ion-exchange is represented by ($[Si_{33}Al_3O_{72}]$ $Cu_{1.5}$). From this formula I have calculated (based on the molar masses of the elements in the unit cell) an amount of 4.226 wt % of Cu in the material. This is the stoichiometric maximum 100% ion-exchange corresponding to a Cu/Al ratio of 0.5.

11. With these correlations, I created the preparation recipes for the different CHA-catalyst samples with the different SAR values and Cu/Al ratios (or Cu and CuO concentrations, respectively). In these calculations, I determined the appropriate amount of Cu-precursor needed (Cu-acetate) to produce the desired Cu/Al ratio via ion exchange given the SAR and amount of zeolite in the ion-exchange slurry.

12. When calculating the Cu/Al ratios of the materials I prepared, I was asked to include only the aluminum from the zeolite and ignore any other aluminum present in the resultant catalyst material, including any aluminum from the binder or other sources.

binder was used in an amount of 12 wt %, resulting in an overall washcoat loading amount of 150 g/L coated catalyst volume. After coating, the substrates were dried and calcined. The catalysts with a SAR of 13 were calcined for 2 hours at 500 °C in air, while the other catalysts were calcined for 4 hours at 640 °C in air.

14. In addition to the Cu-CHA zeolite coated substrates, I was also asked to make a number of copper loaded beta zeolite (BEA) coated substrates. To create these samples, I used a BEA zeolite with a SAR of 30, which I copper loaded using the same procedure described above to produce Cu/Al ratios in the range of approximately 0.15 to 0.55. I then coated substrates with the copper loaded BEA zeolite material in the same manner I describe above for the Cu-CHA materials.

15. Then multiple 1 inch diameter x 3 inch length core samples were drilled out of each Cu-CHA coated substrate to allow for testing. A fresh core sample was retained from each Cu-CHA coated substrate. And, a core sample from each Cu-CHA coated substrate was aged for 50 hours at 800 °C using a forced flow-through of hydrothermal atmosphere containing 10 vol. % of oxygen and 10 vol. % of water vapor balanced by nitrogen. This treatment is assigned as 50 B 800 in the attached exhibits.

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