Study of the oxidation of ammonia in contact with a Ag-exchanged zeolite by DRIFT operando

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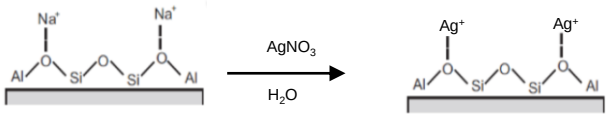
Introduction:

High energy density materials are used as propellants in the aerospace industry. If materials with a higher energy density than hydrazine could be obtained, the orbiting costs would be reduced. Compounds with a high nitrogen content are promising candidates due to the stability of nitrogen as a reaction product. Cyclotriazane (N_3H_3) is particularly interesting due to its ring energy.

Synthesis pathway studied:

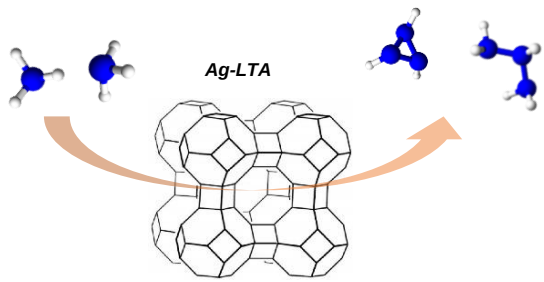
Oxidation of ammonia in contact with Ag-exchanged LTA zeolite
➔ Inspired by Seff and co-worker* who reported the formation of cyclotriazane and triazane (N_3H_5) after NH_3 treatment of an Ag-zeolite.

- **Preparation of Ag-LTA by ion exchange**



* Kim, Y.; Seff, K. *J. Am. Chem. Soc.* **1977**, *99* (21), 7057–7059.

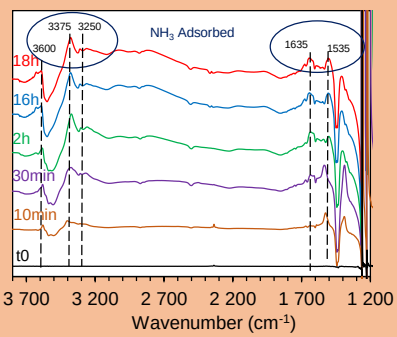
- Contact flow of NH_3 (2000 ppm in N_2) at 40°C
- Thermo-desorption of species confined in the zeolite under He flow



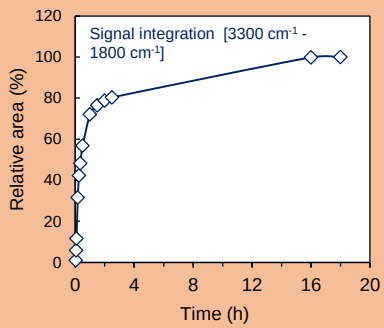
NH_3 adsorption recorded by DRIFT operando:

Na-LTA

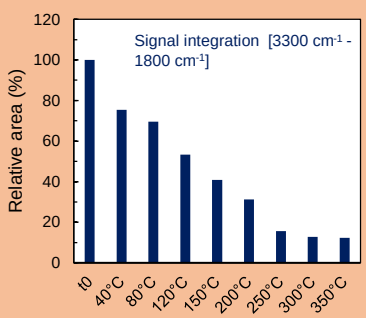
FT-IR spectra



Adsorption Kinetics



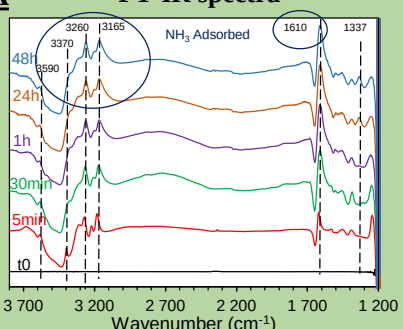
Thermo-desorption



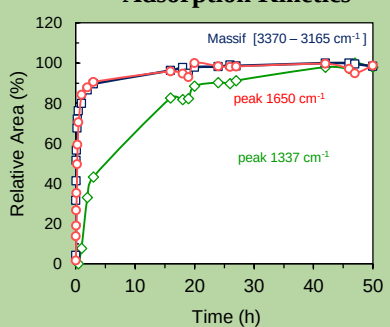
***Observation**: Rapid saturation of the surface by NH_3 / Efficient thermo-desorption of NH_3 on Na-LTA (~12 % remaining after 350°C)*

Ag-LTA

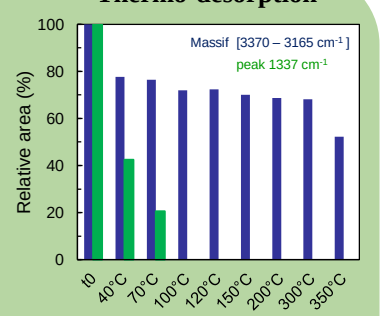
FT-IR spectra



Adsorption Kinetics



Thermo-desorption



***Observation**: Emergence of a new peak at 1337 cm^{-1} with apparition kinetics different from NH_3 adsorption / Signal vanishing after the thermo-desorption step in contrast to strongly adsorbed NH_3 (~52 % remaining after 350 °C)*

Conclusion and perspectives

A DRIFT operando study of the adsorption of NH_3 on an Ag-exchanged zeolite revealed a non-identified species on the surface of the solid. Moreover, a strong affinity between Ag and NH_3 was observed. These results suggest that new species could be formed by the reaction between Ag and NH_3. Currently we are focusing on the isolation and identification of these species.