



Experiment title: Determination of Cu sites structure in Cu-ZSM-5 and Cu-Y zeolites

Experiment number: CH132

Beamline: GILDA D8

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Shifts:
9

Local contact(s):
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Report: Stimulated by the great interest reached by Cu exchanged zeolites in the direct conversion of NO into N₂ and O₂ [1], we have performed a XAFS study of the local Cu environment in Cu^I-Y prepared by an original gas phase reaction of H-Y with CuCl [2] and in Cu^{II}-Y prepared by conventional ion exchange with aqueous cupric salt. XAFS measurements, were performed in transmission mode at GILDA D8 beamline [3] using a double crystal Si(331) monochromator operating in a dynamical **sagittal** focusing mode [4], using an *ad hoc* cell already described elsewhere [5], allowing *in situ* sample activation up to 800 K and gas dosage to be made. The Cu^I-Y sample has been measured in: i) vacuum; ii) after interaction with CO; iii) after interaction with NO; iv) after reduction at 500 K in CO atmosphere for one hour and subsequent evacuation (In Figs. indicated as samples Y, YCO, YNO and *Yrid* respectively). The Cu^{II}-Y sample has been measured only in vacuum conditions (In Figs. indicated as sample Y2). XANES, UV-Vis and IR (NO dosed at 77 K) techniques prove that the copper species in the prepared samples have the desired oxidation state. Reduction of Cu^I-Y with CO leads to the formation of small copper clusters well identified by EXAFS, XANES and UV-Vis spectroscopies, while interaction with NO partially oxydate Cu^I to Cu^{II}. For both as prepared Cu^I-Y and Cu^{II}-Y samples EXAFS does not observe first shell Cu-Cu bond lengths, so excluding the presence of "so called" Cu-dimers in zeolites prepared following our methods. Our results complete previous XAFS data on Cu-Y reported in the literature [6], and will be presented at XAFS IX conference [7]. Analysis on Cu-ZSM-5 samples is in progress. We thank the ESRF Chem. Lab.

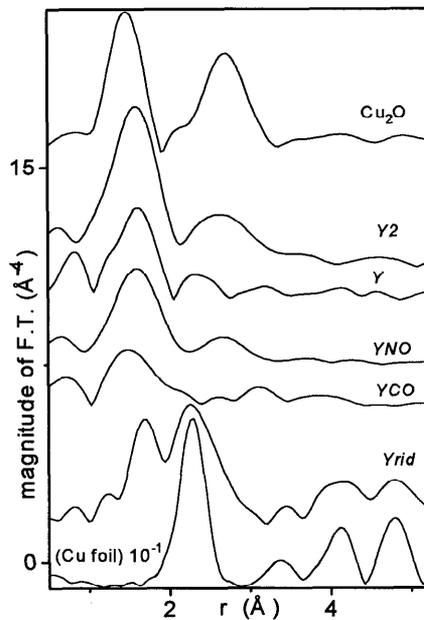
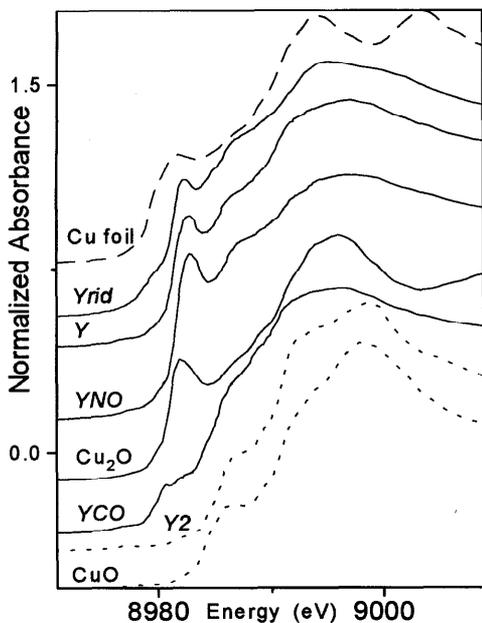


Fig. 1: XANES spectra: dashed, full and dotted curves indicate Cu species having mainly 0, I, II as oxidation state (Cu foil) and 3-13 \AA^{-1} (other samples) range.

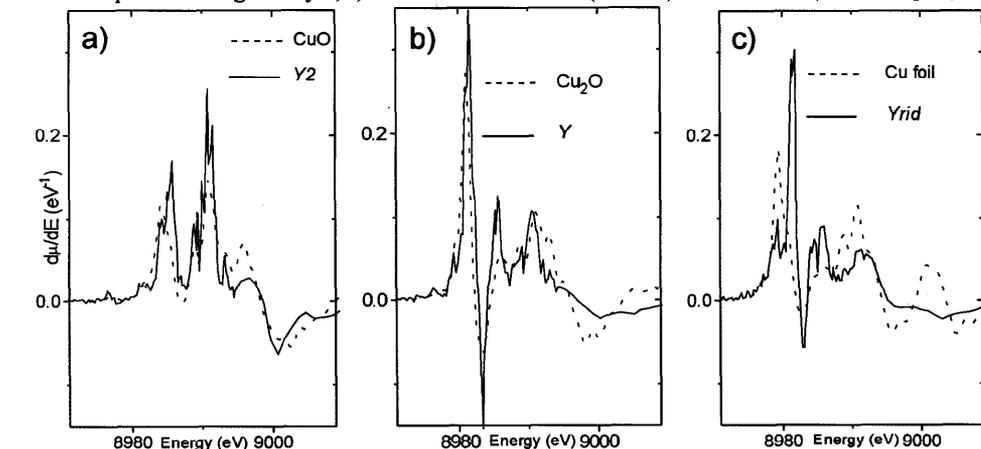


Fig. 3: $d(\mu x)/dE$ spectra of Y2, Y and Yrid (parts a, b and c) compared with spectra of model compounds.

References: [1] Iawamoto M. *et al.*, *Catal. Today* **10** (1991) 57; *Appl. Catal.* **69** (1991) L15; *Stud. Surf. Sci. Catal.* **54** (1990) 121; Li Y. and Hall W. K., *J. Phys. Chem.* **94** (1990) 6145; [2] Spoto G. *et al.*, *Catal. Lett.* **13** (1992) 39; *Appl. Catal. B* **3** (1994) 151; *J. Chem. Soc. Faraday Trans.* **91** (1995) 3285; Amberti C. *et al.*, *J. Phys. Chem.* submitted. [3] Pascarelli S. *et al.*, *ESRF Newsletters* **23** (1995) 17. [4] Pascarelli S. *et al.*, *J. Synch. Rad.* **3** (1996) 147. [5] Bordiga S. *et al.*, *J. Phys. Chem.* **98** (1994) 4125; *Nucl. Instr. Meth. B* **97** (1995) 23; *J. Catal.* **158** (1996) 501. [6] Tanabe S. *et al.*, *J. Chem. Soc. Chem. Commun.* (1989) 875; *Bull. Chem. Soc. Jpn.*, **63** (1990) 192; *J. Phys. Chem.*, **94** (1990) 4207; Yamashita H. *et al.*, *ibid.*, **100** (1996) 397. [7] D'Acapito F. *et al.* *J. Phys. IV (France)*, submitted.