



Experiment title: EXAFS of biomimetic Cu and Fe complexes in aqueous solutions and low concentrations in molecular sieves

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CH-1278

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Report:

Zeolite Y encapsulated Cu(II) histidine complexes show promising catalytic activity in the epoxidation of alkenes with peroxides at relatively low temperatures. Depending on the reaction conditions during the ion exchange reaction in which the complex is brought in the zeolite Y structure, different copper(II) histidine complexes are formed.

In order to be able to make a structure-reactivity relation, we studied the different zeolite Y encaged copper(II) histidine complexes by different characterization techniques. To learn more about the local geometry around the central copper ion, the crystal field strength, site symmetry and the spin nature, we wanted to analyse the different zeolite Y immobilized copper(II) histidine complexes as well as aqueous copper(II) histidine solutions by their EXAFS and their XANES spectra. The results of the aqueous copper histidine complexes are necessary to be able to compare the complexes in the zeolite to the complexes in the ionexchange solution.

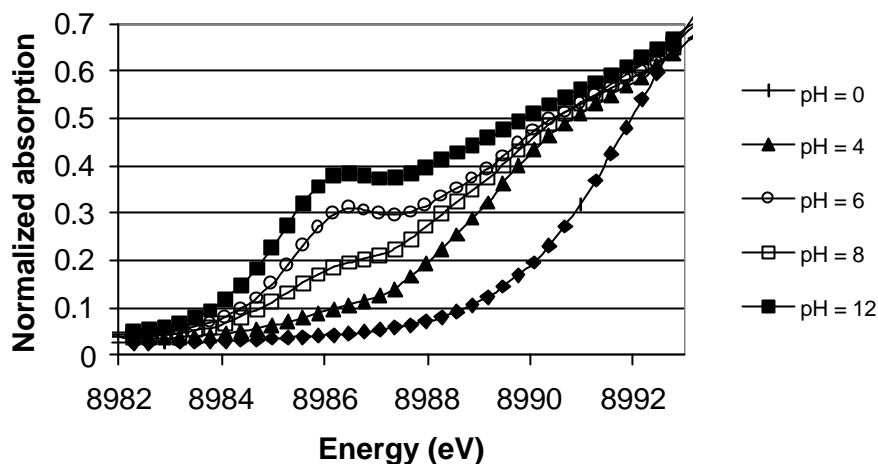


Figure 1: Pre edge features of the aqueous copper(II) histidine complex as function of the solution pH

The pre edge features of the aqueous copper(II) histidine were found to be highly dependent of the pH of the solution (see figure 1). The pre edge feature at 8986.5 eV increases up to a pH value of 6. In the pH region of 6–10 the intensity of the feature decreases, but increases again in the 10-12 pH region. The same feature can be found in the zeolite Y immobilized copper(II) histidine complex (see figure 2). An additional small pre edge feature can be found in the 8983-8984 eV region of the zeolite Y immobilized copper(II) histidine complex.

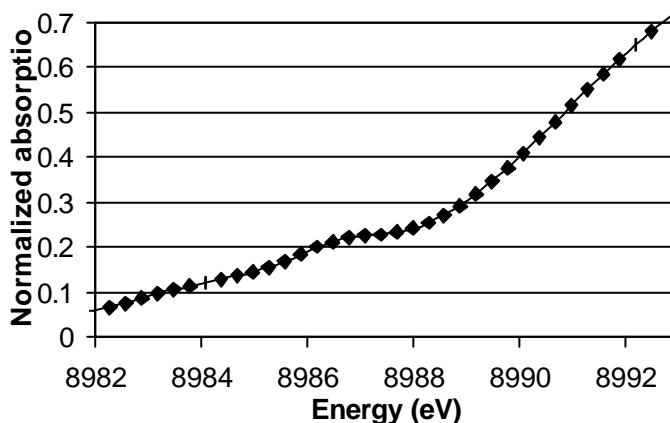


Figure 2: Pre edge features of a zeolite Y immobilized copper(II) histidine complex

The first analyses of the EXAFS results on the aqueous show in the pH range 4–10 the presence of a Jahn-Teller distorted copper histidine complex with four N/O atoms in the equatorial plane at a distance of 1.96Å and two N/O atoms in an axial position at 2.20Å. In the zeolite Y immobilized copper(II) histidine samples also a concentration dependent interaction with the zeolite matrix can be found.