

Project report

CH 1282

In-situ XAS studies on copper zeotypes and zeolites

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Since the establishment of the copper exchanged zeolite CuZSM-5 high NO decomposition activity there has been a great interest in copper materials and their catalytic properties [1-3]. In this project, we wish to study the effects of adsorption of NO on the active metal sites (extraframework and framework) in some copper- and silver-exchanged and –incorporated zeotypic materials by XAS using a pseudo in situ cell.

The as-synthesised and calcined materials have been characterised by XAS at ambient pressure and temperature, but it was important to establish if reduction of copper takes place during the adsorption of NO. The mechanism for selective nitric oxide reduction on conventionally ion-exchanged zeolites is believed to occur via the reduced Cu^{I} form[4]. It is of special interest to compare these results with our ion-exchanged copper zeolite made by a nonconventional procedure, and also copper incorporated zeotypes. Presence of Cu^{I} is easily detectable using XAS due to a strong pre-edge feature in addition to information obtained about the local structures about the target atoms.

The samples were placed in an in-situ cell and calcined at 500°C in Helium for 1 hour before activating the sample in O_2 . The sample was then treated with propene (C_3H_6 , 0.36% in He) and allowed to cool down before collecting XAS spectra. The sample was then reheated to 500°C, first in a NO (0.6% in He) environment and at last in propene. XAS spectra were collected after each treatment to establish the presence of $\text{Cu}^+/\text{Cu}^{2+}$ redox-pairs.

The specific gases are chosen for comparison with deNOx conversion measurements already performed in the Catalysis Laboratory at Nottingham Trent University[5]. The gases are commonly used to simulate real conditions experienced in exhaust from cars [6,7]. Figure 1 show the results for the CuZSM-5 sample made by classic ion-exchange where Cu^{2+} is reduced to Cu^+ after treatment in propene, and then oxidised back to Cu^{2+} after the NO treatment. Finally the treatment with propene reduces Cu^{2+} back to Cu^+ . In the case of the CuZSM-5 made hydrothermally the treatment in NO does not oxidise Cu^+ to Cu^{2+} completely (Fig. 2).

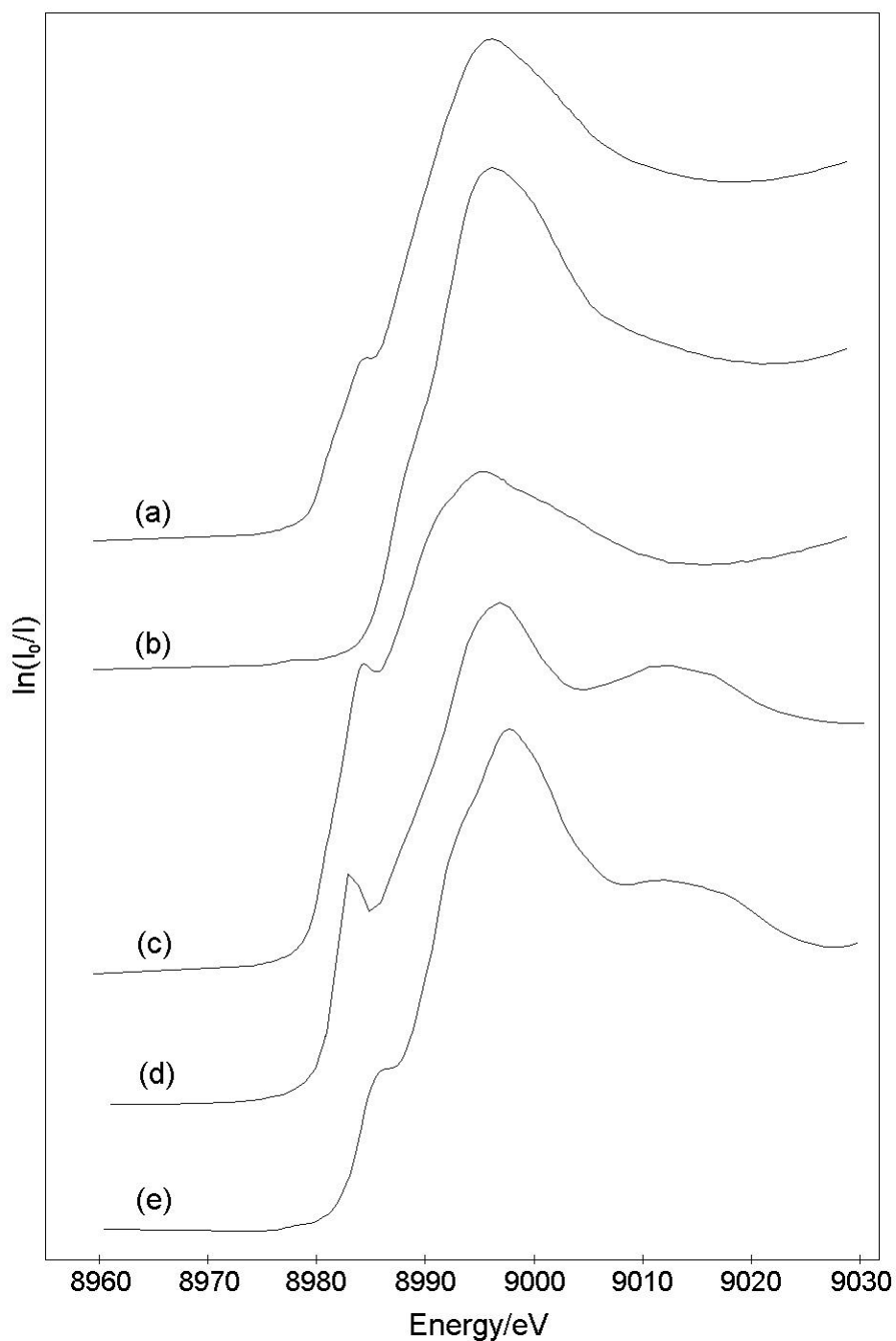


Fig. 1 Normalised Cu K-edge XANES of CuZSM-5 made by classic ion-exchange; (a) calcined and activated in oxygen (6%) before treatment with propene (0.36%); (b) sample from (a) treated with NO (0.6%); (c) sample from (b) treated with propene (0.36%) again; (d) copper(I) oxide; (e) copper(II) oxide.

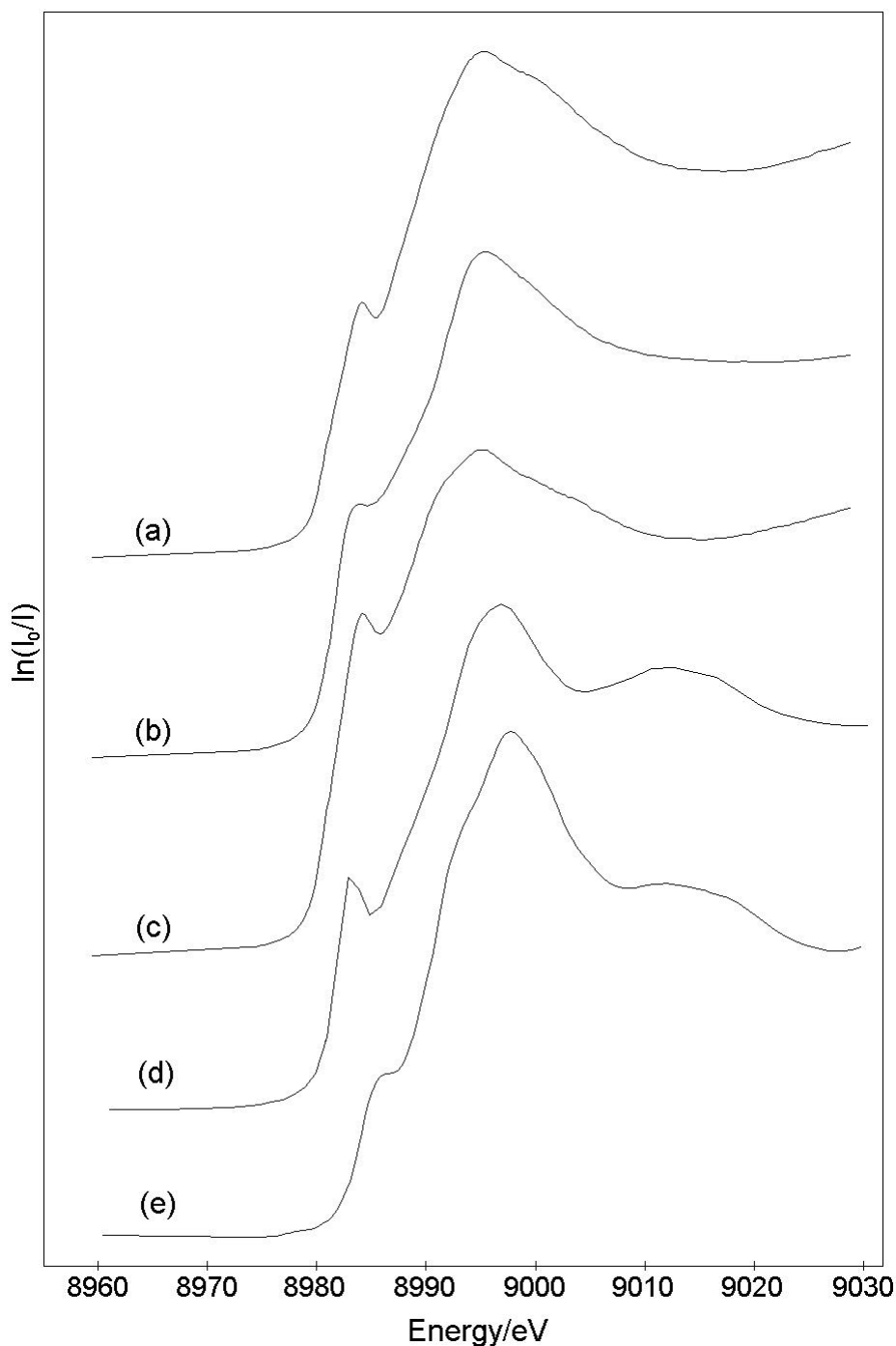


Fig. 2 Normalised Cu K-edge XANES of CuZSM-5 made by hydrothermal ion-exchange; (a) calcined and activated in oxygen (6%) before treatment with propene (0.36%); (b) sample from (a) treated with NO (0.6%); (c) sample from (b) treated with propene (0.36%) again; (d) copper(I) oxide; (e) copper(II) oxide.

References

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