Beamtime Report
SNBL BM01B
01-01-1000, May 2016.

Karina Mathisen[*], Kate McAulay, Said Laassiri, Karsten Kirste*, Tina Kristiansen Voss* and Justin J. S. Hargreaves
*Department of Chemistry, NTNU, Norway
School of Chemistry, University of Glasgow

For this beamtime the proposed deNOx experiment on copper materials could not be performed as crucial reaction gases were not delivered at the scheduled date. We instead chose to focus on a different system and reaction, a continued investigation of the $CoRe_4$ –system for ammonia synthesis with hydrogen and nitrogen since these gases were available. The data were collected at the Re L_{III}- and Co K-edge. The filling mode, 16-bunch, of the ring during these experiments were not optimal and resulted in longer counting times during data collection. In addition we also experienced problems with the beamline equipment, as the blowers malfunctioned in the middle of an experiment and we could not get it restarted until the next morning.

We were, however able to redo some experiments from beamtime 01-01-987, now using stoichiometric reaction gas which was not available for the initially allocated time. We are in this project interested in observing the behaviour of rhenium and cobalt for the ammonia synthesis and the effect of different reducing conditions during pre-treatment. We were, with the new near-operando conditions, able to recreate activity profiles observed in the home lab using the MS during in-situ measurements (Fig. 1).

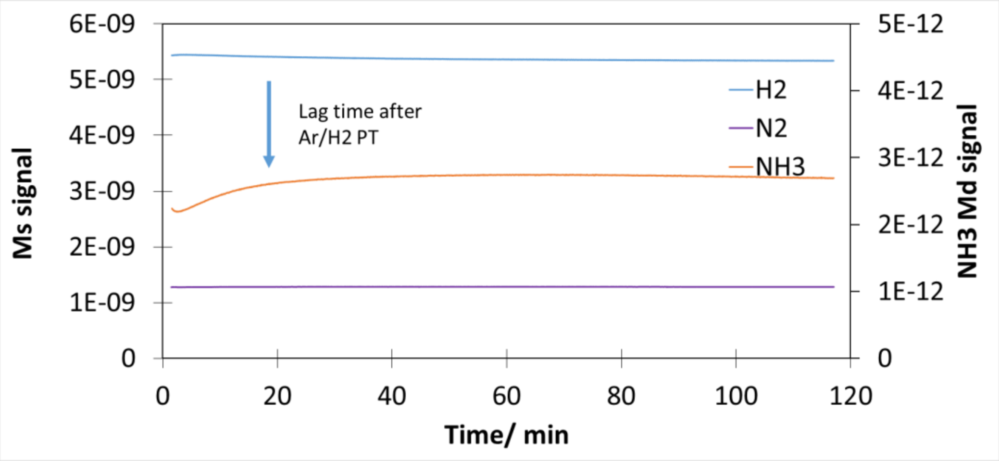

Fig. 1. Lag-time observed for ammonia production following nitrogen free pre-treatment from MS signal during in situ XAS/XRD data collection, corresponding with home lab catalytic results.

These results also helped us understand the role of the different pre-treatments for the formation of the metallic Re-Co pair, and also a possible nitride phase detected in reaction gas with both pre-treatments. This is confirmed from local Re and Co coordination shells from EXAFS analysis. (Fig. 2)

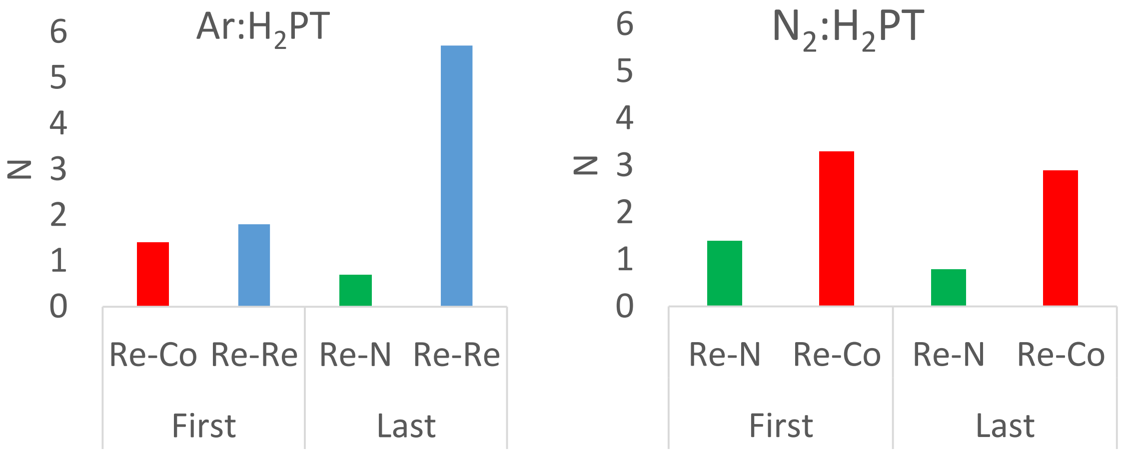

Figure 2: Changes in Re-Re, Re-Co and Re-N absorption pair coordination numbers in reaction gas following the different pre-treatments.

Using a switching valve provided by Dr. van Beek, we were able to monitor ammonia production and local Re and Co environment when either N_2 or H_2 is removed from the system. This revealed that ammonia production continues in hydrogen after the sample has been saturated with reaction gas (Fig.3). This experiment will now we recreated in the home-lab.

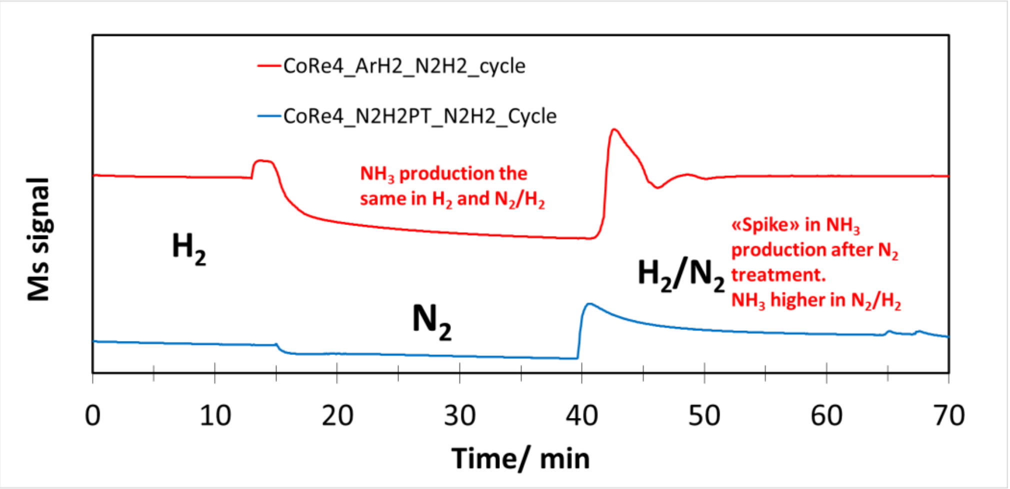

Figure 3: Ammonia production during stepwise switching of reaction components.