Electronic Supplementary Information (ESI)

Ammonia decomposition catalysis using lithium-calcium imide

Joshua W. Makepeace[1,2], Hazel M. A. Hunter[2], Thomas J. Wood[2], Ronald I. Smith[2], Claire A. Murray[3] and William I.F. David[1,2]*

[1]Inorganic Chemistry Laboratory, University of Oxford, South Parks Road, Oxford, OX1 3QR, UK
[2]ISIS Facility, Rutherford Appleton Laboratory, Harwell Campus, Didcot, OX11 0QX, UK
[3]Diamond Light Source Ltd, Rutherford Appleton Laboratory, Harwell Campus, Didcot, OX11 0DE, UK
* email bill.david@stfc.ac.uk

Contents

XRD of synthesised $Li_2Ca(NH)_2$

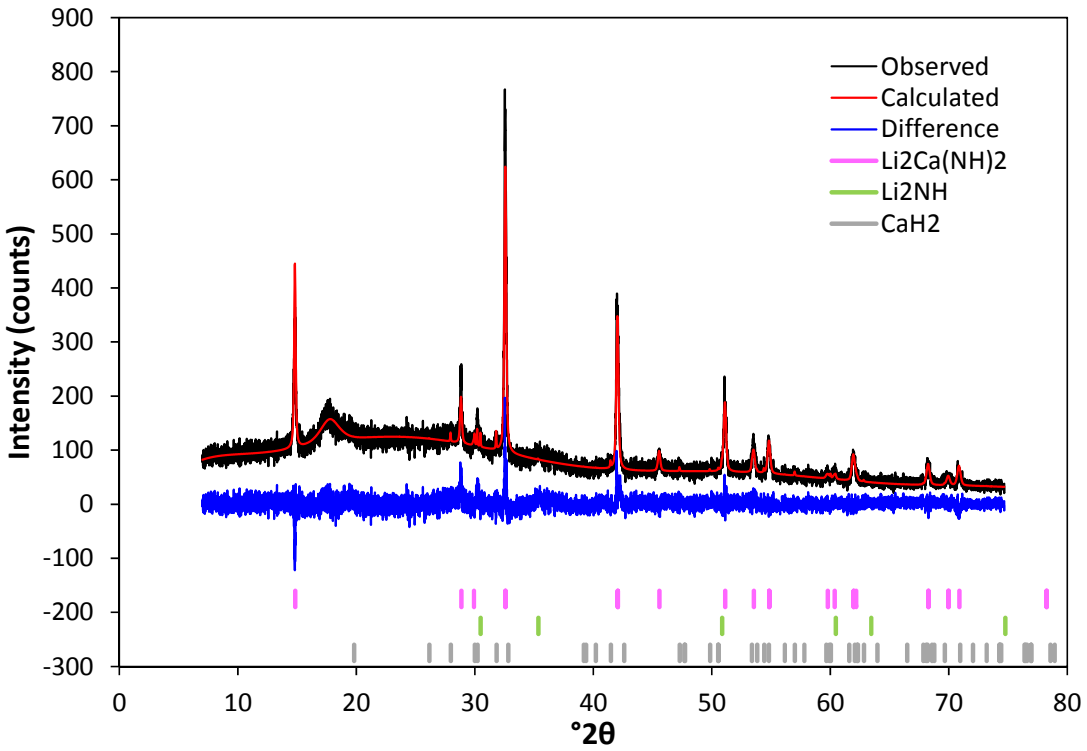


Figure S1: Powder X-ray diffraction pattern of the sample of lithium-calcium imide used in the ammonia decomposition and synchrotron powder diffraction experiments.

XRD of synthesised $Li_2Ca(ND)_2$

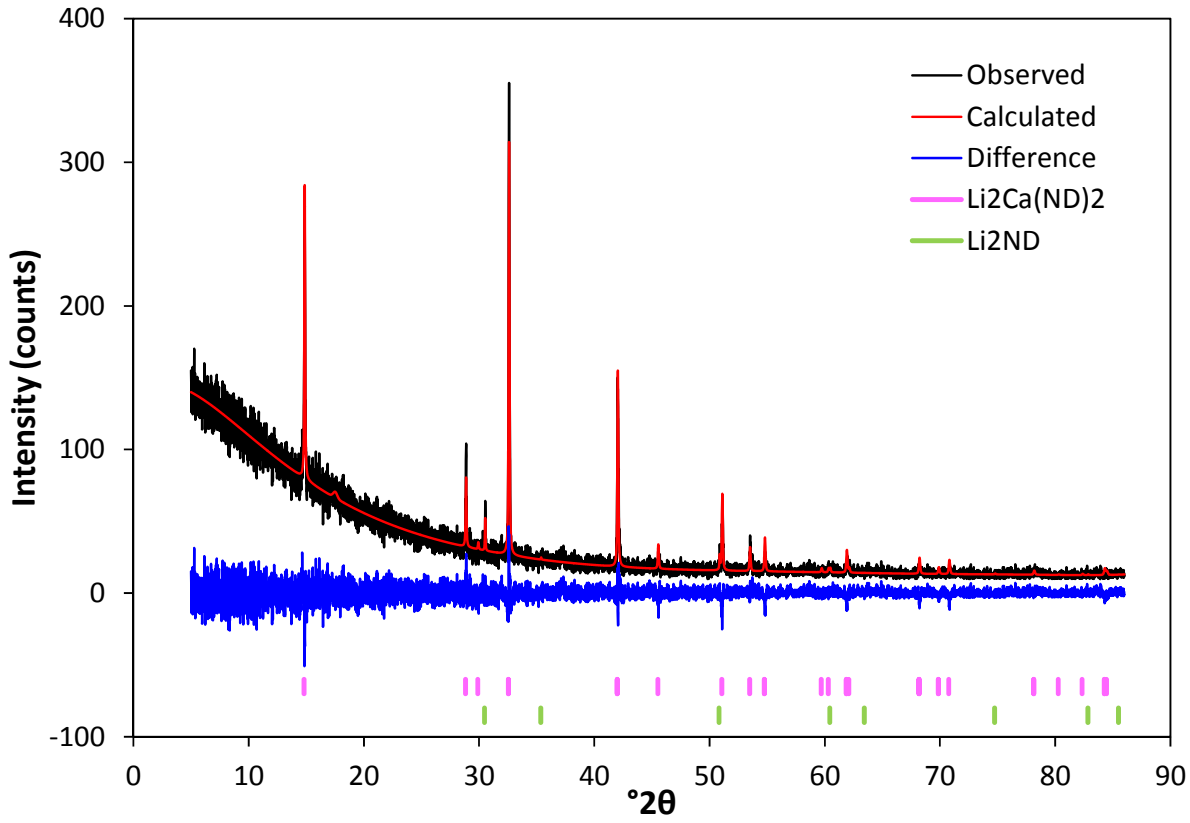


Figure S2: Powder X-ray diffraction pattern of the sample of deuterated lithium-calcium imide used in the neutron powder diffraction experiment.

Ammonia decomposition setup

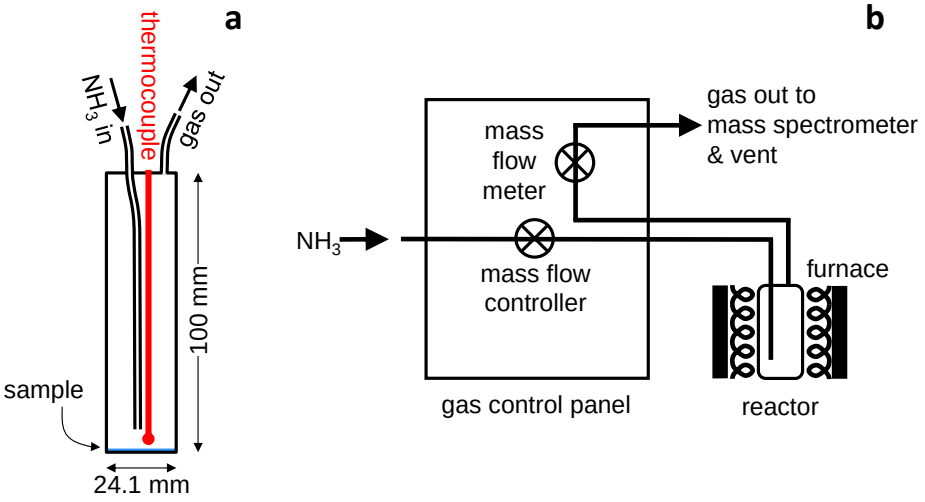
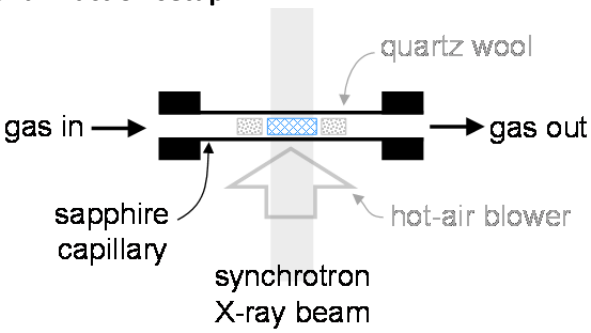


Figure S3: Reactor and experimental setup: (a) a typical 46.9 cm^3 reactor showing NH_3 inlet, gas outlet, and thermocouple temperature probe positions, with a 0.5 g sample, drawn to scale. (b) Experimental gas handling setup, where the inlet NH_3 gas flow is controlled prior to the reactor, and the outlet gas flow is monitored by a mass flow meter and analysed by a mass spectrometer.

Synchrotron X-ray powder diffraction setup



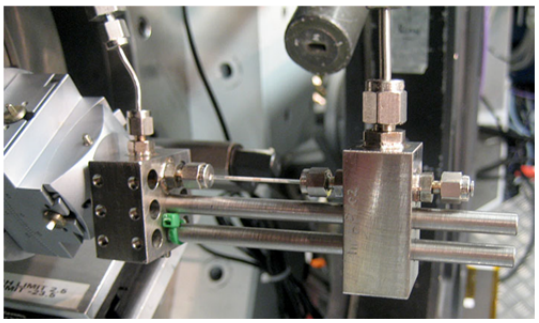


Figure S4: Synchrotron X-ray powder diffraction setup highlighting the powder sample sandwiched between two quartz wool plugs and contained within a 0.9mm i.d. sapphire capillary. Gas flows continuously through the capillary and the sample is heated to the desired temperature using a hot air blower. A photograph of the setup (without the hot air blower) is shown below.

***In-situ* neutron diffraction setup**

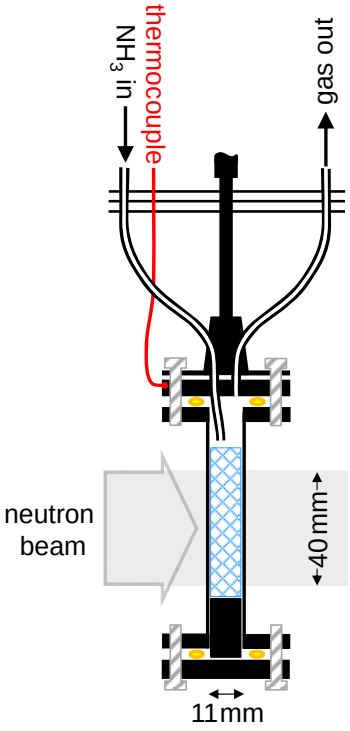


Figure S5: *In-situ* neutron diffraction setup highlighting the powder sample contained within a 0.35mm wall thickness stainless steel cell and held in position in the neutron beam.

XRD of $LiNH_2$ + SiO_2

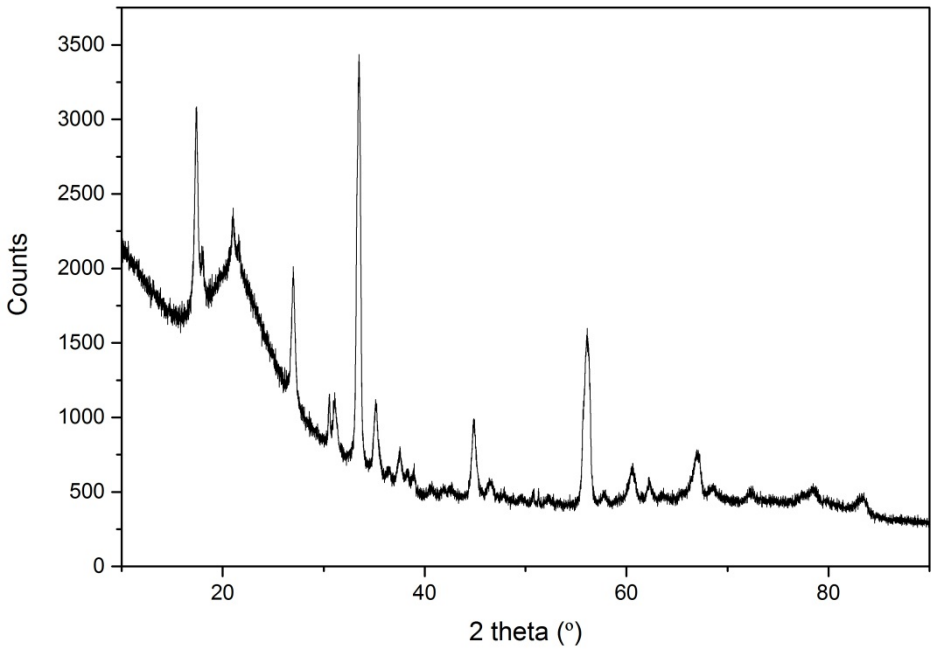


Figure S6: X-ray powder diffraction pattern of the post-reaction sample for $LiNH_2$ + SiO_2 heated under ammonia to 570 °C.

Combined DSC-TGA of $Li_2Ca(NH)_2$

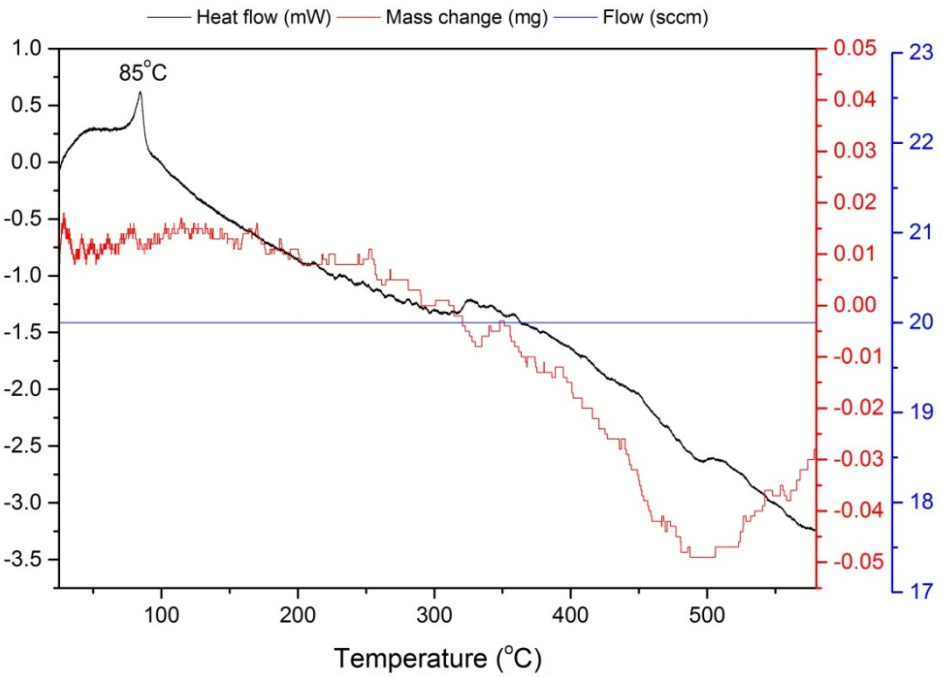


Figure S7: Combined DSC-TGA data for the heating of lithium-calcium imide (5 °C/min) under 20 sccm flowing nitrogen gas.

Synchrotron XRD of $Li_2Ca(NH)_2$ under flowing He

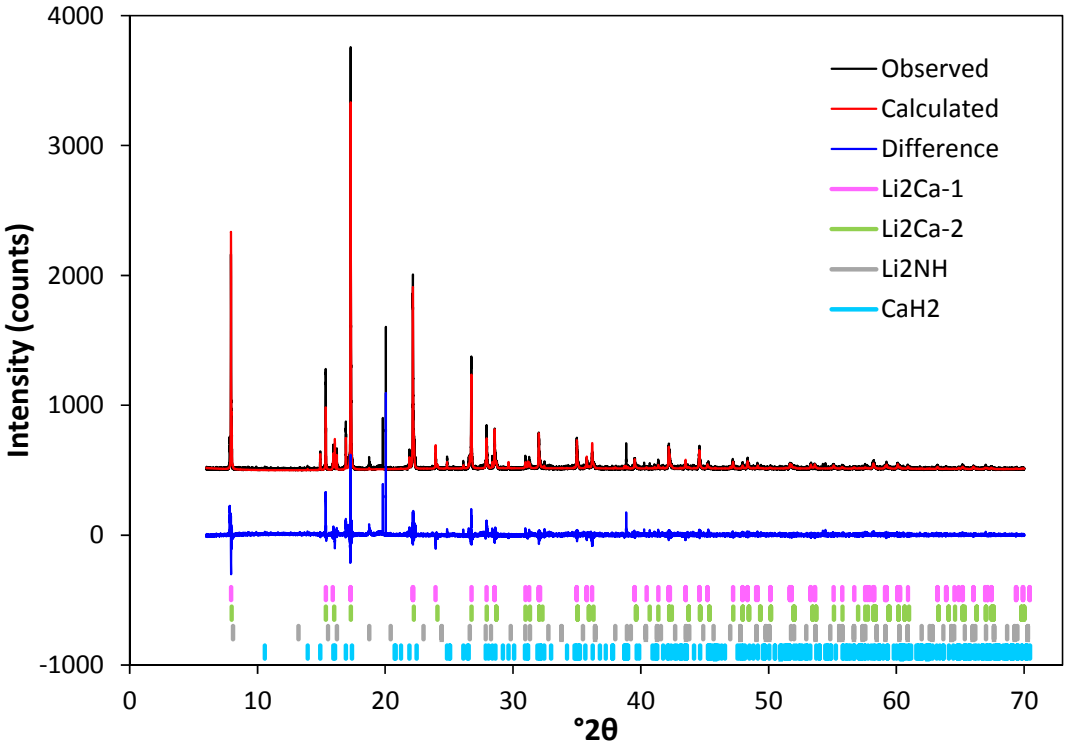


Figure S8: Synchrotron powder X-ray diffraction pattern of the initial sample of lithium-calcium imide used for helium flow experiment.

Synchrotron XRD pattern of $Li_2Ca(NH)_2$ under flowing NH_3

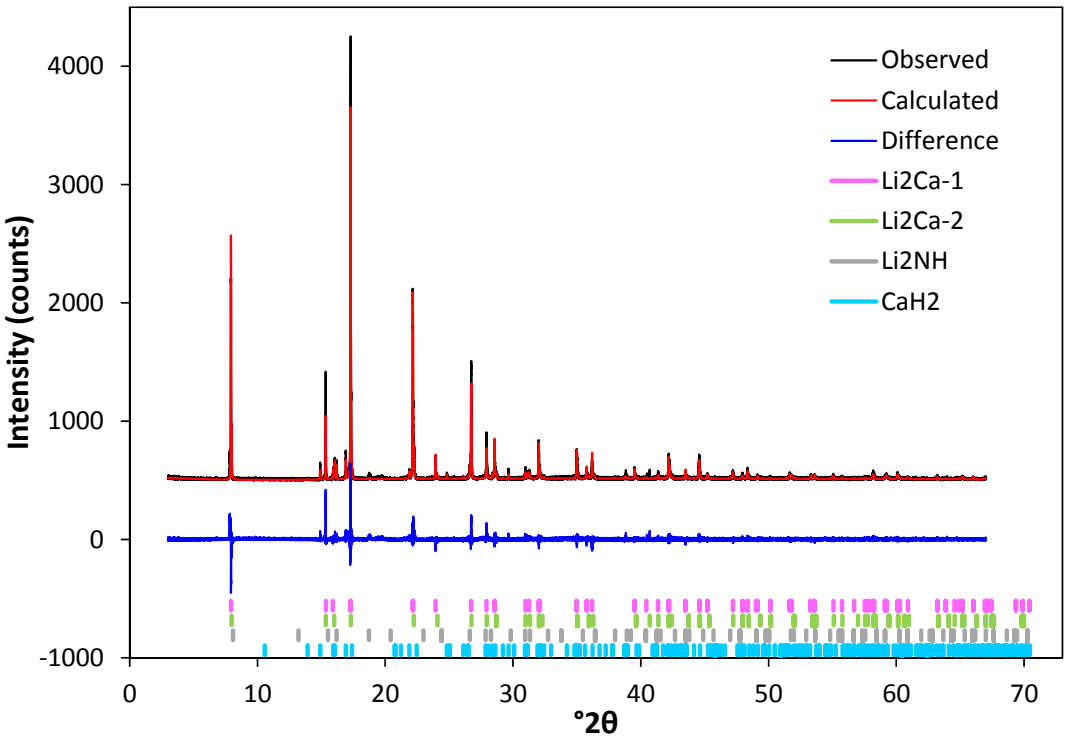


Figure S9: Synchrotron powder X-ray diffraction pattern of the initial sample of lithium-calcium imide used for the ammonia flow experiment.

$Li_2Ca(ND)_2$ lattice parameters from neutron powder diffraction experiment

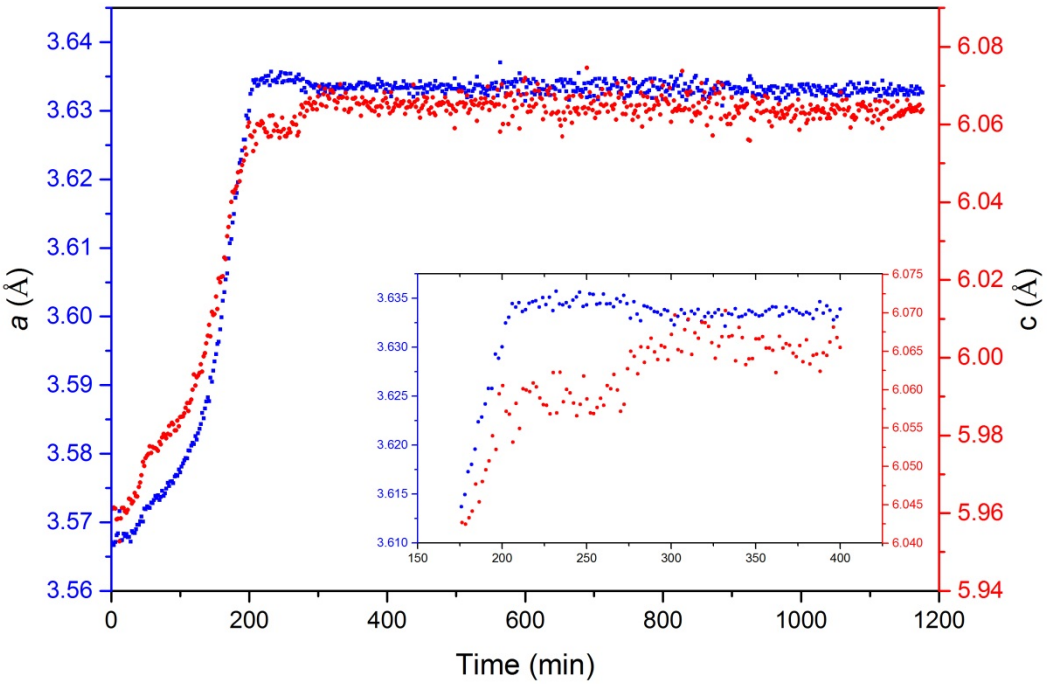


Figure S10: The lattice parameters of $Li_2Ca(ND)_2$ extracted from the Rietveld analysis of neutron diffraction data collected during the *in-situ* experiment, with the period between 180 minutes and 400 minutes enlarged in the inset graph.