Mechanisms and Kinetics of NH_3 Oxidation and NO_X Formation during Fixed-Bed and Fluidised-Bed Combustion of Ammonia

Presented by Samuel Ronald Holden

Centre for Energy (M473), The University of Western Australia, Crawley, WA 6009, Australia

Email: sam.holden@research.uwa.edu.au

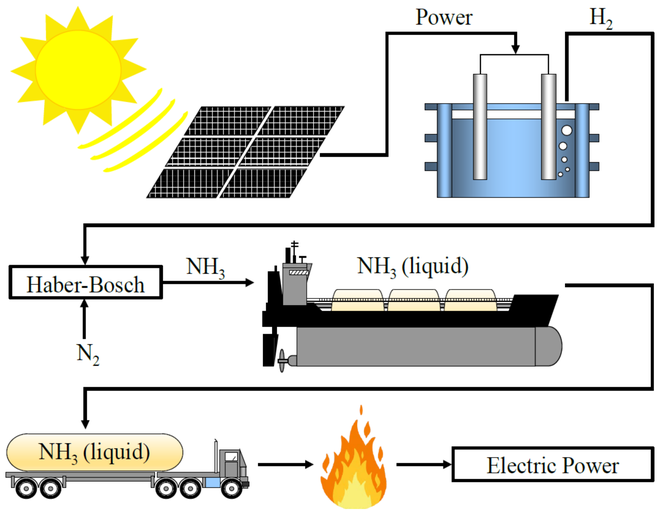
FEnEx CRC THEME

- Program 2: Hydrogen Export & Value Chains

IMPACT AND IMPORTANCE

- We are pioneering the concept and work on NH_3 oxidation / combustion in the presence of solid media
- Ammonia (NH_3) is a carbon-free fuel
- NH_3 is formed by the combination of N_2 and H_2 in the technologically mature Haber-Bosch process
  - Plenty of N_2 from air, free for the time being
  - H_2 can be produced by water electrolysis driven by renewable power
- NH_3 can be easily stored and transported compared to other renewably produced fuels
- NH_3 oxidation / combustion follows:

$$4NH_3 + 3O_2 \rightleftarrows 2N_2 + 6H_2O + (trace\ NO_X) \qquad \Delta H^0 = -46.2\ kJ/mol$$



- Relative to conventional hydrocarbon fuels, ammonia as a fuel has several drawbacks:
  - High autoignition temperature
  - Excessive ignition energy
  - Low flame propagation speed
  - Perceived nitrogen oxides (NO_X) formation and emission

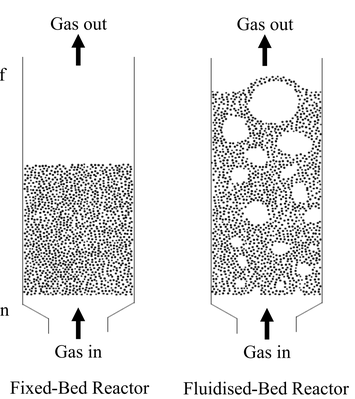
The UWA Centre for Energy has proposed a novel concept of fluidised-bed combustion of ammonia

Fluidised-Bed

- Excellent heat and mass transfer environment
- Expected to achieve the same level of conversion as flow reactors (empty-bed) at lower temperatures

Fixed-Bed

- A special case of a fluidised-bed reactor (flow rate < minimum fluidisation velocity)
- Research tool to provide foundational reaction information



PROJECT AIMS

- Systematic measurement of species profiles as a function of the following parameters:

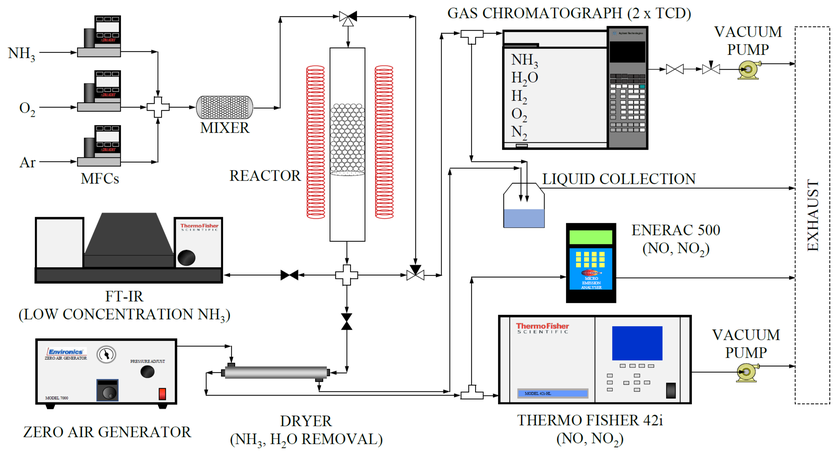
Parameter	Condition
Input Gas Composition	< 8% NH_3
Equivalence Ratio	0.6, 0.8, 1.0, 1.2
Reaction Temperature	1000-1400K
Contact Time	< 2 seconds
Bed Material Type	Quartz, alumina, zirconia
Bed Material Size	0.8-0.9mm, 1.2-1.43mm, 2.36-2.5mm

- Determination of ammonia oxidation kinetics / mechanism
- Understanding of NO_X formation / destruction

RESEARCH METHODOLOGY

Experimental

- Ar replaces N_2 in air for combustion of NH_3, allowing nitrogen-containing product species to be traced
- Bed material: quartz (amorphous) has been selected as the primary bed material (voidage ~0.58)
- Flow rate: < 6 SLPM at STP
- Pressure: atmospheric
- Temperature: < 1150°C
- All lines post reactor are maintained > 100°C with heating tape to best avoid condensation



- Arrhenius Equation is the first step for determination of the overall oxidation reaction based on experimental results:

$$R = k[NH_3][O_2]$$

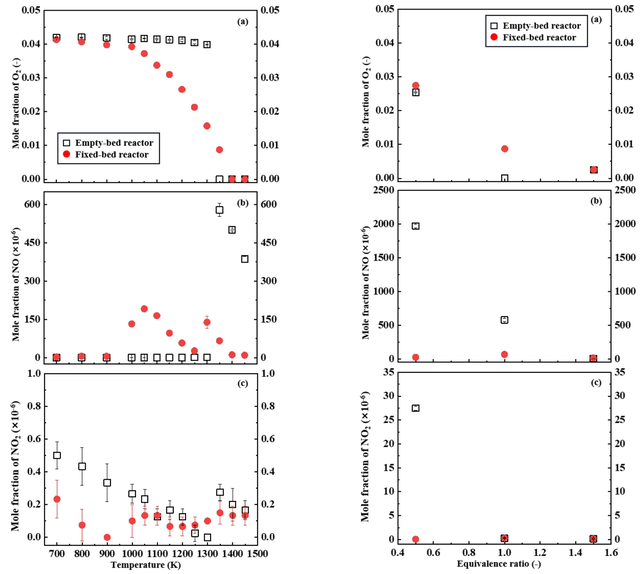
$$ln(k) = ln(k_0) - \frac{E}{RT}$$

where R is the reaction rate; k the rate constant; and $[NH_3]$ and $[O_2]$ the concentration of NH_3 and O_2 respectively; k_0 the pre-exponential factor; E the activation energy; and T the temperature

Computational Modelling

- Ansys Chemkin-Pro – models the chemical reactions
- Ansys Fluent – models the heat / mass transfer
- Chemkin model based on Nakamura, Otomo, and Stagni mechanisms
- Modify existing mechanisms in Ansys Chemkin-Pro for development of a 'new mechanism' to best represent NH_3 fixed-bed oxidation

PRELIMINARY RESULTS



The quartz fixed-bed, in comparison to the empty-bed, leads to:

- Increased reaction conversion at lower temperatures (stoichiometric equivalence ratio)
- Reduced NO_X formation
  - Across a range of temperatures
  - For stoichiometric and lean equivalence ratios

ANTICIPATED OUTCOMES

- Knowledge
  - NH_3 oxidation kinetics
  - NH_3 oxidation mechanism
- Technology
  - Lab-scale fixed- and fluidised-bed NH_3 oxidation / combustion reactor
- Emissions
  - NO_X formation / destruction mechanism in ammonia oxidation / combustion
  - Able to incorporate SCR or SNCR downstream of reactor if NO_X emissions remain unacceptably high

PROJECT RESEARCHERS

- Samuel Ronald Holden – PhD Scholar, Centre for Energy, The University of Western Australia
- Zhezi Zhang – Laboratory Manager, Centre for Energy, The University of Western Australia
- Dongke Zhang – Centre for Energy, The University of Western Australia
- Yin Wu – General Manager / CTO, Shanxi Keteng Environmental Protection Technology Co. Ltd.
- Yinsheng Niu – Director, Technical Centre, Shanxi Keteng Environmental Protection Technology Co. Ltd.

ACKNOWLEDGEMENT

This work receives financial support from the Australian Research Council under the Discovery Projects Scheme (DP210103766) and the Future Energy Export CRC (FEnEx CRC Project # 21.RP2.0059). S.R. Holden receives a full PhD scholarship from the Australian Government.

PROJECT PARTNERS

Centre for Energy

"energy for today and tomorrow"