The role of the Mars – van Krevelen mechanism in the synthesis of ammonia with metal nitride catalysts

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Abstract:
The development of the Haber Bosch Process was a landmark achievement of the 20th Century which can be directly credited with the sustainment of a significant proportion of the global population through access to synthetic fertiliser. On a global scale, the process produces ca 180 million tonnes of ammonia per annum with an annual projected growth rate of 1.5-3.0% to meet growing demand. The entire process, including the generation of feedstock, is responsible for ca. 2% of global energy demand and ca 1.6% of anthropogenic CO_2 emissions. There is therefore great interest, and potential, in the identification of novel, more sustainable, strategies towards ammonia synthesis – these are directed towards the development of smaller scale and more localised, reduced scale processes. Such processes could be achieved through the development of electrocatalytic routes to take advantage of sustainable electricity production, photocatalytic routes and the development of more conventional heterogeneous catalytic approaches operational under less severe process conditions (the Haber Bosch Process currently operates at ca 400°C and > 100 atmospheres pressure with a promoted iron catalyst – the conditions applied are dictated by the requirement to achieve acceptable process conditions, with the reaction being favoured thermodynamically at lower reaction temperatures).

In this presentation, the possibility of the development of metal nitride based catalysts operational through the Mars - van Krevelen mechanism wherein the lattice nitrogen is reactive will be outlined. A combination of experimental and computational modelling data will be presented to support this suggested pathway. The possible utility of this general approach for the preparation of other large scale products will also be outlined.