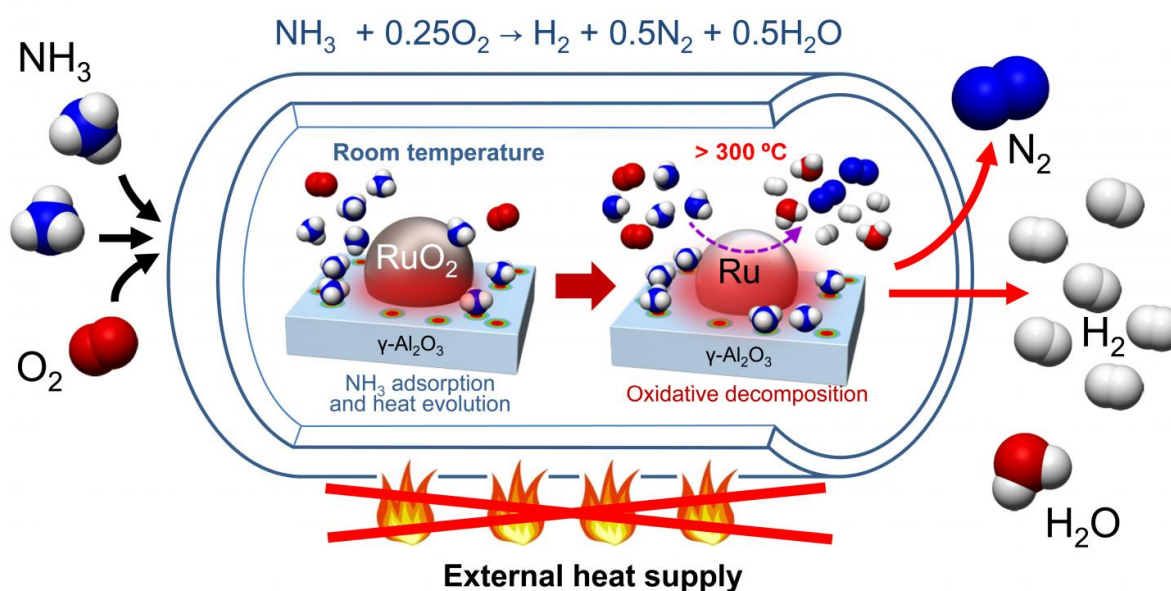


Catalyst for the carbon-free production of hydrogen gas from ammonia

May 19 2017, by Heather Zeiger



Credit: Katsutoshi Nagaoka

(Phys.org)—Hydrogen has the potential to provide an alternative, clean energy source, particularly as applied to fuel cell technology. Current fuel sources involve carbon-containing fossil fuels or carbon-containing organic molecules, which result in the production of excess CO₂, a greenhouse gas. Several initiatives, including a national initiative in

Japan, seek to create a low-carbon usage society by using alternative fuel sources.

The Energy Carriers initiative in Japan is a national project that is specifically looking at ways to efficiently store and transport hydrogen. One way to do this is to use ammonia as a hydrogen source. However, discovery of an efficient process for breaking down ammonia has proved difficult, largely because the catalytic process to break down ammonia requires the continuous addition of heat, which can be prohibitively expensive.

Katsutoshi Nagaoka, Takaaki Eboshi, Yuma Takeishi, Ryo Tasaki, Kyoto Honda, Kazuya Imamura, and Katsutoshi Sato of Oita University in Japan have developed a method using a novel [catalyst](#) for producing hydrogen from ammonia without the addition of external heat through the catalytic cycle. Their work appears in *Science Advances*.

The decomposition of ammonia into hydrogen and nitrogen is an endothermic process, meaning that it requires the addition of energy to obtain products. This means that traditional catalytic decomposition reactions require the addition of a large amount of heat to obtain a useful amount of hydrogen gas.

Nagaoka et al. developed a catalyst that is made of a RuO_2 nanoparticle supported on $\gamma\text{-Al}_2\text{O}_3$ catalyst bed. After purging their catalyst of H_2O and CO_2 , ammonia and oxygen were added to the reaction vessel where ammonia was adsorbed onto the catalytic surface, resulting in an increase in temperature. This increase in temperature catalyzed the oxidative decomposition of ammonia, an exothermic process. This heated up the reaction, which in turn, provided the energy for the endothermic decomposition of ammonia into hydrogen and nitrogen.

The catalyst pre-treatment did require heating to remove water and

carbon dioxide, but it did not require subsequent re-heating. Tests on catalyst cycling showed that after the initial pre-treatment of the $\text{RuO}_2/\gamma\text{-Al}_2\text{O}_3$ catalyst with helium at 300°C , the catalyst was able to cycle three times and still produce hydrogen in maximum yields. Furthermore, these studies included oxidative passivation to ensure that no heat was produced from oxidation of Ru to RuO_2 . In practice, oxidative passivation will not be necessary. So, even though heating is required to pre-treat the catalyst, heating is not required for additional cycles of the catalyst.

In an effort to understand how the $\text{RuO}_2/\gamma\text{-Al}_2\text{O}_3$ catalyst works, Nagaoka et al. compared the maximum catalytic bed temperature that results from self-heating of $\text{RuO}_2/\gamma\text{-Al}_2\text{O}_3$ to $\text{RuO}_2/\text{La}_2\text{O}_3$, a known ammonia decomposition catalyst. They found that the aluminum-based catalyst heated to a maximum temperature of 97°C , while the lanthanum-based catalyst heated to a maximum temperature of 53°C . This is important because the auto-ignition temperature for the oxidative combustion of ammonia is 90°C , and it explains why better reaction yields were seen with $\text{RuO}_2/\gamma\text{-Al}_2\text{O}_3$.

The authors point out that this difference in adsorption temperature is likely due to the favorable interaction between ammonia, a basic molecule, and Al_2O_3 , which is a Lewis acid. La_2O_3 , on the other hand, is a Lewis base.

Additionally, the authors looked at the difference between using bare $\gamma\text{-Al}_2\text{O}_3$ as a catalyst and $\text{RuO}_2/\gamma\text{-Al}_2\text{O}_3$. They found that 90% of the ammonia adsorbs onto bare $\gamma\text{-Al}_2\text{O}_3$ compared to the catalyst bed and the RuO_2 nanoparticle. This implies that ammonia is chemisorbed onto the nanoparticle and $\gamma\text{-Al}_2\text{O}_3$, which then promotes multilayer physisorption.

Overall, this type of catalyst is helpful in providing enough heat to

overcome the needed heat requirements for the endothermic decomposition of ammonia into hydrogen and nitrogen gas. This study shows that self-heating catalysis is a viable option for exploring solutions to the practical difficulties in using [ammonia](#) as a [hydrogen](#) fuel source.

More information: Katsutoshi Nagaoka et al. Carbon-free H₂ production from ammonia triggered at room temperature with an acidic RuO₂/γ-Al₂O₃ catalyst, *Science Advances* (2017). [DOI: 10.1126/sciadv.1602747](#)

Abstract

Ammonia has been suggested as a carbon-free hydrogen source, but a convenient method for producing hydrogen from ammonia with rapid initiation has not been developed. Ideally, this method would require no external energy input. We demonstrate hydrogen production by exposing ammonia and O₂ at room temperature to an acidic RuO₂/γ-Al₂O₃ catalyst. Because adsorption of ammonia onto the catalyst is exothermic, the catalyst bed is rapidly heated to the catalytic ammonia autoignition temperature, and subsequent oxidative decomposition of ammonia produces hydrogen. A differential calorimeter combined with a volumetric gas adsorption analyzer revealed a large quantity of heat evolved both with chemisorption of ammonia onto RuO₂ and acidic sites on the γ-Al₂O₃ and with physisorption of multiple ammonia molecules.

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