Catalysis of N₂O Decomposition: Harvesting Energy from Nitrogen Waste

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Introduction

Nitrous oxide (N₂O) is a greenhouse gas with a global warming potential 310 times
dthat of CO₂, but it also has the potential to release usable energy as it
decomposes into harmless N₂ and O₂.  With the help of an efficient and robust
catalyst, N₂O emissions from such sources as wastewater treatment plants and
hospitals can be broken down to produce electrical power for those facilities, or
stored for use as a monopropellant.  Additionally, a wastewater recycling system
that harvests energy would be useful on long space missions.

Purpose and Hypothesis

The goal of this study is to characterize the performance and learn the
mechanics of catalysts that can help decompose N₂O, in order to find an energy-
efficient catalyst that is both cost-effective and robust in the presence of
contaminants such as O₂ and water vapor.  Rhodium oxide (Rh₂O₃) is expected to be
efficient (require little energy input) to fully decompose N₂O) but
expensive, whereas iron oxide (Fe₂O₃) is expected to be less efficient for pure
N₂O, but less expensive and less effected by contaminants.  Ideally, this study
will prove that Fe₂O₃ is sufficiently energy-efficient to be an affordable catalyst
for hospitals and water treatment plants looking to implement this technology.

Materials and Methods

Materials that are likely candidates as catalysts, mainly metal oxides, will be
analyzed both experimentally and in simulation.

A catalyst bed has been constructed with two gas lines as input so that catalyst
performances can be measured for various gas mixtures.  Gas samples are either
taken from the exit of the catalyst bed or from one of the sample ports along its
length, so the length of the catalyst bed is also a variable to be tested.  Heat tape
is wrapped around the catalyst bed and covered in insulation so that the
percentage N₂O decomposition can be measured over a range of temperatures.
A more efficient catalyst will require less heat input for full decomposition.

Simultaneously, Rh₂O₃ and Fe₂O₃ crystals have been simulated using the
density functional theory (DFT) software package, GPAW.  N₂O molecules are
being simulated close to various surfaces of these crystals so that adsorption
energies and reaction rates can be calculated.  Crystal surfaces that show
promise in simulation can then be tested experimentally.

Results

Tests to date show that Rh₂O₃’s catalytic performance is not significantly
affected by the presence of either N₂ or O₂ in the gas mixture.  In the case of
O₂, this result is surprising because theory would indicate that O₂ binds to the
same sites on the catalyst as N₂O, so O₂ was expected to interfere with
decomposition.

Computational work so far has found lattice constants and band gaps for Rh₂O₃
and Fe₂O₃ consistent with experimental results, demonstrating the software’s
reliability and providing optimized crystal structures that are now in simulation
with N₂O molecules in various orientations.  The band gap was expected to be
low using PBE, since DFT typically underestimates band gaps, which is why
methods exist to correct for this, such as the DFT-U extension.

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Table 1: Computational results compared with experimental results

<table>
<thead>
<tr>
<th>Temperature, °C</th>
<th>N₂O/N₂ Mixture</th>
<th>% N₂O, Exit Flow</th>
<th>% N₂O, Exit Flow</th>
<th>% N₂O, Exit Flow</th>
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<tbody>
<tr>
<td>200</td>
<td>4% N₂O/96% N₂</td>
<td>2%</td>
<td>2%</td>
<td>2%</td>
</tr>
<tr>
<td>205</td>
<td>4% N₂O/96% N₂</td>
<td>2%</td>
<td>2%</td>
<td>2%</td>
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<tr>
<td>210</td>
<td>4% N₂O/96% N₂</td>
<td>2%</td>
<td>2%</td>
<td>2%</td>
</tr>
<tr>
<td>215</td>
<td>4% N₂O/96% N₂</td>
<td>2%</td>
<td>2%</td>
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</table>

Table 2: Summary of computational and experimental results

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Value</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>Temperature, °C</td>
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<td>200</td>
<td>200</td>
</tr>
<tr>
<td>N₂O/N₂ Mixture</td>
<td>4% N₂O/96% N₂</td>
<td>4% N₂O/96% N₂</td>
<td>4% N₂O/96% N₂</td>
</tr>
<tr>
<td>% N₂O, Exit Flow</td>
<td>2%</td>
<td>2%</td>
<td>2%</td>
</tr>
</tbody>
</table>

Conclusions

Rhodium oxide has proven a more robust catalyst than originally expected, and
simulated catalysts line up with experimental data so far, so the ongoing
simulations with gas molecules are expected to accurately identify which sites
are most active in catalysis.

Acknowledgments

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References

2. [V.S. Khvostunova et al.]: “Mechanistic origins of the different activity of Ru 2004-5 and Fe 2004-5 in N2O

Bibliography

Further Information

Feel free to direct questions and/or comments to Ashley Hicks at
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