

3 Results

All of the results shown in this section are based on an operating cell potential of 0.7 V, current density of 5000 A/m², and inlet temperature of 500 °C (773 K) of supply gases. Figure 2 shows the ammonia mole fraction at the anode. Ammonia enters the anode channel at 1 atm. Within the anode domain, it decomposes into hydrogen and nitrogen and as a result its concentration decreases both in the direction of bulk flow and also in the direction anode-to-electrolyte. Note that the lowest mole fraction of ammonia is 0.83 indicating a low conversion rate.

Figure 3 shows the corresponding plot for hydrogen mole fraction. The inlet gas has zero hydrogen. But the hydrogen concentration increases as it is produced in the anode, but simultaneously decreases as it is consumed in the electrochemical reactions. At 0.7 V, the maximum hydrogen mole fraction is 0.0864. If there were no electrochemical reactions occurring at the anode/electrolyte interface, the maximum hydrogen mole fraction would have been higher.

Figure 4 shows the steady state temperature distribution. Cathode and anode gases enter at 773 K. For these conditions, the changes in temperature are not large. It decreases to a minimum value of 770.3 K and increases to a maximum of 774.2 K. This temperature sink is due to the endothermic ammonia decomposition reaction. The temperature rise is due to heat produced by the fuel cell reactions and ohmic heating. As a result, there is an initial sink at the anode entrance region, followed by a gradual rise in temperature, consistent with previous works [32,33].

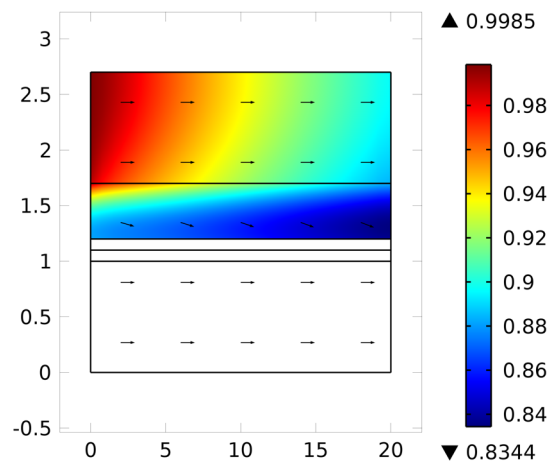


Figure 2. Ammonia Mole Fraction.

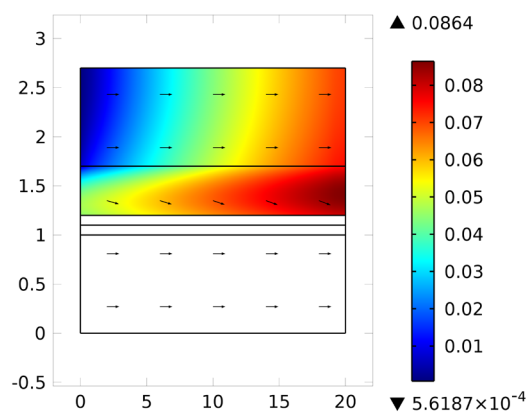


Figure 3. Hydrogen Mole Fraction.

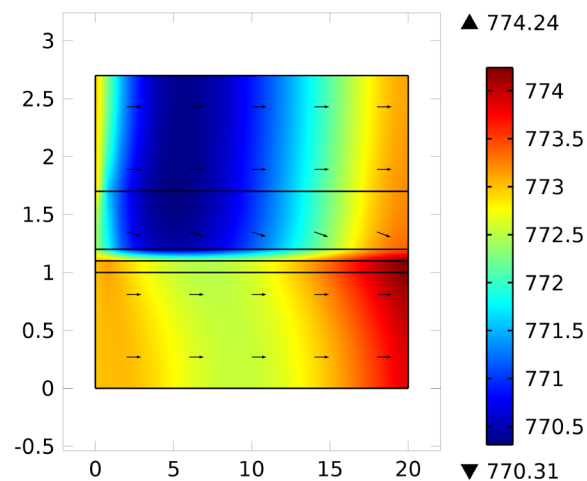


Figure 4. Temperature Distribution (units: K).

4 Discussion

The results presented in the previous section are based on an operating cell potential of 0.7 V, a current density of 5000 A/m². It should be noted that in our previous model of a high temperature ammonia-fed SOFC [33], the cell potential was 0.45 V with a current density of 3940 A/m². This means that the present intermediate temperature SOFC produces a power density of 3500 W/m², compared to 1773 W/m² for the high temperature SOFC. The intermediate temperature SOFC produces nearly twice as much power as the equivalent high temperature SOFC primarily because of the high conductivity of the proton conducting electrolyte.

Figure 2 showed that the ammonia utilization is only 17% at the temperature used in this work. This is compared to 100% in the previous high temperature model [33]. These results demonstrate that even though the rate of ammonia decomposition decreased with temperature, the fuel cell performance was not compromised at intermediate temperature operation.

The lower rate of ammonia decomposition is attributed to two factors: reduced kinetic rates as well as hydrogen inhibition occurring at the lower temperature. This conversion can be improved by recycling the anode exhaust as shown in our previous work [33], as well as pressurizing the ammonia. However this would also exacerbate the temperature sink.

In high temperature SOFCs, this temperature sink is much more pronounced. In previous models for high temperature ammonia-fed SOFCs, temperature sinks of 67 °C have been reported [32,33], which pose deleterious material problems. In this work, the temperature sink is only 3 °C. This is primarily due to the reduced rate of ammonia decomposition. The results shown in this work suggest that problems of thermal shock can be significantly reduced by operating ammonia-fed SOFCs at intermediate temperatures.

5 Conclusion

This work reported the first model of an intermediate temperature ammonia-fed SOFC in the literature. It took into account hydrogen inhibition using Temkin-Pyzhev kinetics. Because of the reduced temperature and the phenomenon of hydrogen inhibition, the rate of ammonia decomposition was lower than high temperature SOFCs, however the fuel cell performance was not compromised. In fact, the results have shown that maintaining a high electrolyte conductivity offsets the effects of reduced ammonia decomposition rates at the lower temperature.

The results have shown that the temperature sink (also known as the “cold spot”) associated with the endothermic ammonia decomposition reaction was significantly mitigated at intermediate temperature operation primarily because of the reduced rate of ammonia decomposition.

In future publications, the author plans to further develop modelling of intermediate temperature SOFCs to include comparisons between Tamaru and Temkin-Pyzhev kinetics at various temperatures, and also to consider a combination of both models at the critical temperature range.

References

1. C. Zamfirescu and I. Dincer, “Using ammonia as a sustainable fuel,” *J. Power Sources*, vol. 185, pp. 459–465, 2008.
2. J.O. Jensen, A.P. Vestbo, Q. Li and N.J. Bjerrum, “Development of a high pressure microbalance for hydrogen storage materials”, *J. Alloys Compd.* Vol. 446-447, pp. 723–728, 2007.
3. L.M. Zhang, Y. Cong, W.S. Yang and L.W. Lin, “The energy efficiency of onboard hydrogen storage”, *Chinese J. Catalysis*, vol. 28, pp. 749–751, 2007.
4. L. Pelletier, A. McFarlan and N. Maffei, “Ammonia fuel cell using doped barium cerate proton conducting solid electrolytes,”, *J. Power Sources*, vol. 145, pp. 262–265, 2005.
5. N. Maffei, L. Pelletier, J.P. Charland and A. McFarlan, “Direct ammonia fuel cell using barium cerate proton conducting electrolyte”, *J. Power Sources*, vol. 140, pp. 264–267, 2005.
6. Q.L. Ma, R.R. Peng, L.Z. Tian and G.Y. Meng, “Direct utilization of ammonia in intermediate-temperature solid oxide fuel cells”, *Electrochem. Comm.* Vol. 8, pp. 1791–1795, 2006.
7. D. Cheddie, “Ammonia as a hydrogen source for fuel cells: a review,” in: D. Minic, Ed., *Hydrogen Energy – Challenges and Perspectives*, InTech Publishers, 2012, pp. 333-362.
8. R. Lan, J.T.S. Irvine and S. Tao, “Ammonia and related chemicals as potential indirect hydrogen storage materials”, *Int. J. Hydrogen Energy*, vol. 37, pp. 1482 -1494, 2012.
9. T.V. Choudhary, C. Svadinaragana and D.W. Goodman, “Catalytic ammonia decomposition: COx-free hydrogen production for fuel cell applications”, *Catal. Lett.* Vol. 72, pp. 197, 2001.
10. S.F. Yin, Q.H. Zhang, B.Q. Xu, W.X. Zhu, C.F. Ng and C.T. Au, “Investigation on the catalysis of COx-free hydrogen generation from ammonia”, *J. Catal.* Vol. 224, pp. 384, 2004.
11. S.F. Yin, B.Q. Xu, S.J. Wang, C.F. Ng and C.T. Au, “MgO-CNTs (Magnesia-carbon nanotubes) nanocomposites: Novel support of Ru catalysts for the generation of COx free hydrogen from ammonia”, *Catal. Lett.* Vol. 96, pp. 113, 2004.
12. S.J. Wang, S.F. Yin, L. Li, B.Q. Xu, C.F. Ng and C.T. Au, “Investigation on modification of Ru-CNTs catalysts for the generation of COx free hydrogen from ammonia”, *Appl. Catal. B: Env.* Vol. 52, pp. 287, 2004.
13. H.C. Liu, H. Wang, J.H. Shen, Y. Sun and Z.M. Liu, “Preparation, characterization and activities of the nano-sized Ni/SBA-15 catalyst for producing COx-free hydrogen from ammonia”, *App. Catal. A: Gen.* vol. 337, pp. 138–147, 2008.
14. X.K. Li, W.J. Ji, J. Zhao, S.J. Wang and C.T. Au, “Ammonia decomposition over Ru and Ni catalysts supported on fumed SiO₂, MCM-41, and SBA-15”, *J. Catal.* Vol. 236, pp. 181–189, 2005.
15. J. Zhang, H.Y. Xu, X.L. Jin, Q.J. Ge and W.Z. Li, “Characterizations and activities of the nano-sized Ni/Al₂O₃ and Ni/La-Al₂O₃ catalysts for NH₃ decomposition”, *App. Catal. A*, vol. 290, pp. 87–96, 2005.
16. W.Q. Zheng., J. Zhang, Q.J. Ge, H.Y. Xu and W.Z. Li, “Effects of CeO₂ addition on Ni/Al₂O₃ catalysts for the reaction of ammonia decomposition to hydrogen”, *App. Catal. B: Env.* vol. 80, pp. 98, 2008.
17. C. Plana, S. Armenise, A. Monzón and E. García-Bordejé, “Ni on alumina-coated cordierite monoliths for in situ generation of CO-free H₂ from ammonia”, *J. Catalysis*, vol. 275, pp. 228–235, 2010.
18. Y. Lin, R. Ran, Y. Guo, W. Zhou, R. Cai, J. Wang and Z. Shao, “Proton conducting fuel cells operating on hydrogen, ammonia and hydrazine at intermediate temperatures”, *Int. J. Hyd. En.*, Vol. 35, pp. 2637, 2010.
19. A.S. Chellappa, C.M. Fisher and W.J. Thomson, “Ammonia decomposition kinetics of Ni-Pt/Al₂O₃ for PEM fuel cell applications”, *App. Catal. A: Gen.* vol. 227, pp. 231–240, 2002.
20. K. Tamaru, “A ‘new’ general mechanism of ammonia synthesis and decomposition on transition metals”, *Acc. Chem. Research*, vol. 21, pp. 88-94, 1988.

21. S.A. Vilekar, I. Fishtik and R. Datta, "The Peculiar Catalytic Sequence of the Ammonia Decomposition Reaction and its Steady-State Kinetics", *Chem. Eng. Sci.* vol. 71, pp. 333-344, 2012.
22. J. Zhang, H. Xu and W. Li, "Kinetic study of NH₃ decomposition over Ni nanoparticles: The role of La promoter, structure sensitivity and compensation effect", *App. Catal. A: Gen.* vol. 296, pp. 257-267, 2005.
23. S.H. Israni, B.K.R. Nair and M.P. Harold, "Hydrogen generation and purification in a composite Pd hollow fiber membrane reactor: experiments and modeling", *Cat. Today* vol. 139, pp. 299-311, 2009.
24. A. Di Carlo, A. Dell'Era and Z. Del Prete, "3D simulation of hydrogen production by ammonia decomposition in a catalytic membrane reactor", *Int. J. Hydrogen Energy*, vol. 36, pp. 11815-11824, 2011.
25. M. Ni, D.Y.C. Leung and M.K.H. Leung, "Thermodynamic analysis of ammonia fed solid oxide fuel cells: Comparison between proton conducting electrolyte and oxygen ion conducting electrolyte", *J. Power Sources*, vol. 183, pp. 682-686, 2008.
26. E. Baniasadi and I. Dincer, "Energy and exergy analyses of a combined ammonia-fed solid oxide fuel cell system for vehicular applications", *Int. J. Hydrogen Energy*, vol. 36 pp. 11128-11136, 2011.
27. S.A. Hajimolana, M.A. Hussain, W.M.A. Wan Daud and M.H. Chakrabarti, "Dynamic modelling and sensitivity analysis of a tubular SOFC fuelled with NH₃ as a possible replacement for H₂", *Chem. Eng. Res. and Des.*, vol. 90, pp.1871-1882, 2012.
28. M. Ni, D.Y.C. Leung and M.K.H. Leung, "Electrochemical modelling of ammonia-fed solid oxide fuel cells based on proton conducting electrolyte", *J. Power Sources*, vol. 183, pp. 687-692, 2008.
29. M. Ni, D.Y.C. Leung and M.K.H. Leung, "Mathematical modeling of ammonia-fed solid oxide fuel cells with different electrolytes", *Int. J. Hydrogen Energy*, vol. 33, pp. 5765-5772, 2008.
30. M. Ni, D.Y.C. Leung and M.K.H. Leung, "An improved electrochemical model for the NH₃ fed proton conducting solid oxide fuel cells at intermediate temperatures", *J. Power Sources*, vol. 185, pp. 233-240, 2008.
31. F. Ishak, I. Dincer and C. Zamfirescu, "Thermodynamic analysis of ammonia-fed solid oxide fuel cells", *J. Power Sources*, vol. 202, pp. 157-165, 2012.
32. M. Ni, "Thermo-electrochemical modelling of ammonia-fueled solid oxide fuel cells considering ammonia thermal decomposition in the anode", *Int. J. Hydrogen Energy*, vol. 36, pp. 3153-3166, 2011.
33. D. Cheddie, "Modelling of ammonia-fed solid oxide fuel cells," in: A. Mendez-Vilas (Ed.), *Materials and Processes for Energy: Communicating Current Research and Technological Developments*, Formatex Research Center, 2013, pp. 504-511.
34. D. Cheddie, "Modelling the Hydrogen Inhibition Effect on Ammonia Decomposition", *Journal of Energy and Power Engineering*, vol. 8, pp. 662-669, 2014.
35. R. Suranwarangkul, E. Croiset, M.W.Fowler, P.L. Douglas, E. Entchev and M.A. Douglas, "Performance comparison of Fick's, dusty gas and Stefan-Maxwell models to predict concentration overpotentials of a SOFC anode", *J. Power Sources*, vol. 122, pp. 9-18, 2003.

Nomenclature

Table 1. Abbreviations.

Abbreviation	Meaning
SOFC	Solid oxide fuel cell
NO _x	Nitrogen oxides
YSZ	Yttria-stabilized zirconia
SDC	Samaria-doped ceria

Table 2. Symbols.

Symbol	Meaning
β	Permeability
η	Overpotential
μ	Dynamic viscosity
φ	Electrolyte phase potential
ρ	Density
c	Molar concentration
c_p	Specific heat capacity
D	Gas pair diffusivity
F	Faraday constant
H	Enthalpy
j	Current density
j_0	Exchange current density
k	Thermal conductivity
M	Molecular mass
N	Molar flow rate
p	Partial pressure
r	Reaction rate
R	Universal gas constant
S	Source term
s	Entropy
T	Temperature
u	Fluid velocity
x	Mole fraction

Table 3. Subscripts and superscripts.

Abbreviation	Meaning
i,j,k	Chemical species
a, c	Anode, cathode
e	Electrolyte phase
act	Activation
ref	Reference
eff	Effective
T	Thermal