13th U.S. National Combustion Meeting Organized by the Central States Section of the Combustion Institute March 19–22, 2022 College Station, Texas

Swiss-roll Heat Recirculating Ammonia Reformer for Gas Turbine Applications

Patryk Radyjowski^{1*}, Patharapong Bhuripanyo², Chien-Hua Chen¹, and Paul Ronney²

¹Advanced Cooling Technologies, Inc., Lancaster, PA, USA ²University of Southern California, Los Angeles, CA, USA ^{*}Corresponding Author Email: <u>patryk.radyjowski@1-act.com</u>

Abstract: Ammonia (NH₃) is an attractive carbon-free energy carrier alternative to hydrogen with substantially easier storage requirements and higher volumetric energy density. Consequently, NH₃ has been studied as a fuel in the oncoming decarbonized energy economy. A potential application for NH₃ is the ammonia-fueled gas turbine for aviation or electricity generation (the latter as a peaker or emergency plant to generate high-value electricity). However, the inherently low flame speeds of NH₃-Air mixtures result in a narrow flammability range and combustion instabilities, which make ammonia challenging to implement in conventional gas turbines. One potential solution is to reform part of the ammonia into hydrogen and add it to the main ammonia fuel stream to increase the flame speed. With this motivation, a novel Swiss-roll style heat-recirculating fuel reformer is proposed as an upstream fuel cracker for an ammonia-fueled gas turbine system. The Swiss-roll geometry effectively recuperates and transfers the thermal enthalpy from the exhaust stream back into reactants, which enables superadiabatic reaction temperatures in the central reaction core. This elevated temperature accelerates chemical reactions, thus causing the reactants to proceed more quickly toward the chemical equilibrium state. In addition to the kinetic benefit, the superadiabatic reaction temperature provides the thermodynamic benefit by enabling ultra-rich NH₃ partial oxidation. Consequently, substantial amounts of hydrogen can be generated. The hydrogen-rich product stream would then be combined with the neat ammonia supply providing a fuel-air mixture with higher flame speeds compatible with gas turbine applications. The idea has been investigated numerically via 0D Perfectly Stirred Reactor (PSR) and 2D Computational Fluid Dynamics (CFD) studies at elevated pressures. Furthermore, an initial experimental approach has been performed, where an optimized Swiss-roll design, additively manufactured in Inconel 718, was tested under different flow rate conditions. The produced reformate has been characterized and investigated as a fuel additive for ammonia-operated gas turbines.

Keywords: ammonia combustion, gas turbine, fuel reforming, heat recirculation

1. Introduction

The movement towards net-zero emissions has highlighted the need for power generation technologies utilizing carbon-free fuels such as hydrogen (H₂). However, ammonia (NH₃) is a promising energy source for future power generation or used as an H₂ carrier. Ammonia's relatively high volumetric energy density (15.6 MJ/L) is significantly higher than difficult-to-handle cryogenic H₂ (9.1 MJ/L) and nearly three times that of compressed H₂ (5.6 MJ/L). Combined with acceptable gravimetric energy density (22.5MJ/kg), relative ease of production, transport, and storage makes it a favorable candidate as a fuel for the next generation of green gas turbines for aviation and power generation [1]. The net-zero carbon economic analysis by Cesaro

Sub Topic: Novel Combustion Techniques

et al. [2] shows that ammonia-based gas turbine energy generation offers the most economic value for low-capacity factor operation characteristic of peaking power plants. Furthermore, NH₃ is currently in use commercially as a commodity chemical, refrigerant, and fertilizer. Hence the infrastructure for its production, transportation, and distribution is already established.

Despite desirable economic characteristics, direct air-ammonia combustion in gas turbines has proven challenging to implement. Several technical factors contribute to troubled ammonia implementation in gas turbine systems. First, the chemical reaction rates of ammonia combustion are relatively slow, leading to substantially lower laminar flame speed than common hydrocarbon fuels. Hence a much larger combustor is required for a self-sustaining Damköhler number range. Meanwhile, the lower volumetric heat release than hydrocarbon fuels requires higher flow rates, further intensifying the slow reaction rates issue. Low laminar flame speed combined with reduced residence time dictated by the lower calorific value of ammonia leads to significant flame-holding issues. Additionally, the adiabatic flame temperature is usually 100-200 K cooler than popular natural gas fuel. This is important for meeting the turbine inlet temperature requirements upon mixing combustion products with secondary air. Finally, NH₃-air combustion emits substantially more NO_x pollution than current state-of-the-art hydrocarbon burners. Despite the listed difficulties, there are ongoing efforts to develop direct ammonia combustors for gas turbines, such as NH₃-CH₄-H₂ fuel mixing [3], swirl-stabilized burners [4], or rich-quench-lean (RQL) staged combustion approaches [5].

The current study concentrated on an alternative method, where NH₃ is pre-cracked upstream of the gas turbine. In such case, the dedicated reforming device can be optimized for sustaining ammonia chemistry without the need for extensive modifications to the gas turbine itself. The resulting reformate can be combusted directly in an H₂-optimized gas turbine or mixed with supplemental NH₃ to increase energy density, Figure 1A, while retaining the benefit of improved flame speed by blending with H₂ [1,6]. The control over mixing H₂-rich reformate and NH₃ allow fine-tuning of the mixture combustion characteristics, as represented by laminar flame velocity in Figure 1B [7]. Experiments performed by the Valera-Medina [8] identified 70-30% NH₃-H₂ blend as optimal for stable combustion in gas turbines. More recently, SINTEF Energy reported a wide operating range for NH₃-reformate mixtures in unmodified Siemens SGT-750 gas turbines [9]. In both cases, stable operation was achieved with the 70-30% fuel blend, even though direct NH₃ combustion was impossible. Additionally, the reformate introduces a well-incorporated water vapor, which slows down the HNO kinetics, a major NO_x precursor in the NH₃-air oxidation path [10,11].





Sub Topic: Novel Combustion Techniques

Most methods considered for NH₃ reforming are based on thermodynamics, where the ultimate H₂ output is limited by either kinetic or chemical equilibrium, given a sufficient residence time. A summary of chemical equilibria for a range of temperatures and pressure is shown in Figure 2. The high operating pressure required for integration with gas turbines makes low-temperature catalyst-based solutions thermodynamically limited in conversion efficiency, as shown in Figure 2A. Therefore this work studies the use of a novel heat-recirculating Swiss-roll non-catalytic thermal reformer as an ammonia pre-cracker for a gas turbine application. Such a device has no inherent pressure limitations as the conditions in the reaction site are usually sustained above 1000 K.



Figure 2: Chemical equilibrium for: (A) $NH_3 \leftrightarrow H_2 + N_2$ reaction at range of temperatures and pressures [12] (B) $NH_3 + air \leftrightarrow H_2O + H_2 + N_2$ for a range of equivalence ratios Φ ; arrows show predicted H_2 molar fraction



Figure 3: (*A*) Schematic diagram of the Swiss-roll reformer. (*B*) Temperature profile for reaction with and without heat-recirculation.

The concept of a heat-recirculating combustor was originally proposed by Weinberg [13,14]. The Swiss-roll configuration is a combustion chamber inside a spiral heat exchanger, shown schematically in Figure 3A. The cold reactants (fuel-rich mixtures of NH₃ and air) flow into the center reaction zone, where the partial oxidation reforming process occurs, while the hot products (reformate) flow out from the center through the adjacent outlet channel. The heat transfer from the hotter outlet channel to the cooler inlet channel through the spiral heat exchanger significantly increases the reactants' temperature, thus combustion temperature and reaction rates. Figure 3B further illustrates the temperature increase resulting from heat recirculation. Without heat recirculation, the reaction zone temperature is increased by both chemical reaction and heat recirculation. The resulting reaction temperature can be higher than the adiabatic flame temperature, called "superadiabatic" flame temperature. This extends the flammable range of a

variety of fuels and enables the achievement of equilibrium state concentrations. The anticipated operating range for the ammonia reformer is in the ultra-rich regime, where the thermodynamic equilibrium predicts H_2 yields of up to 50% molar fraction, as shown in Figure 2B.

2. Methods

The presented study encompasses numerical modeling and experimental examination of a Swissroll ammonia reformer. The modeling work concentrates on a near full-scale 1ft device, sized to integrate with a MW_{th} power range gas turbine. It is complemented by a small-scale experimental approach on a 6in sub-scale device.

Modeling Work

The modeling approach utilizes a 0-dimensional perfectly-stirred reactor (PSR) combined with a 2-dimensional study Computational Fluid Dynamics (CFD) study of Swiss-roll geometry performance. The PSR calculations determine the important parameters affecting the reforming yield over a range of temperatures and residence times. A 0D adiabatic PSR is simulated via CHEMKIN employing GRI-Mech III detailed chemistry model [15]. The CFD study explores the fuel-reforming capabilities of Inconel-718 Swiss-roll heat-recirculating combustors. The model setup includes the effects of temperature-dependent gas and solid properties. The viscous effects are modeled using Reynolds Stress Model (RSM). Surface-to-surface radiation was modeled via discrete ordinates with an Inconel wall emissivity of 0.35. GRI-Mech III, Otomo's and Song's models [16] were used for the ammonia-air chemistry model, generally producing agreeable results. The outermost surface heat loss to ambient (300K) is modeled via natural convection with a 10 W/m²K heat transfer coefficient and 0.8 external emissivity. The out-of-plane volumetric heat sink's thermal resistance model, as shown in Figure 4, mimics the typical experimental insulation configuration. The inlet boundary is a fixed mass-flow inlet with ambient temperature reactants. The calculations were carried out with Ansys Fluent, and the out-of-plane heat loss model was applied via a user-defined function (UDF). Reasonable measures were taken to ensure grid/timestep-independence results, and conservation of mass, species, and energy were verified for all results.



Figure 4: (Left) Swiss-roll base geometry and (Right) 1-dimensional heat loss resistance model.

The baseline geometry is a 1-ft scale, 3.5 turns cylindrical combustor with 1.25 cm channel width, 0.25 cm wall thickness, and 9.25 cm core diameter. A finned-wall variation is also included in the study, with the fin thickness comparable to that of the wall's, and the heights were scaled such that the effective flow's channel width becomes uniform. The combustor has a single inlet and a single outlet. The inlet boundary condition is a fixed mass-flow inlet, and the outlet is a specified pressure of 10 atm to represent conditions required to interface with gas turbine.

Experimental Work

The experiment aims to establish the operating range, measure the reforming efficiency and validate the numerical model at pressures of 1 bar and 10 bar. A sub-scale 6in outer diameter Swiss-roll reformer has been 3D printed in Inconel 718. Inconel was selected to maximize the operating temperature while maintaining sufficient strength for elevated pressure operation in the future. A 2.5-turn swirl heat exchanger comprises 2.5 mm wide channels separated by 1.5 mm wide walls. The core section is 4in wide and 6in tall, with a profiled inlet at the bottom and a simple outlet at the top. The preheated reactants enter radially and are diverted into an axial direction by a flow straightener. This increases the residence time and helps to stabilize the ammonia combustion in the core. The block diagram of supporting equipment is presented in Figure 5A, while a cross-sectional view of the Swiss-roll device is shown in Figure 5B. The extensive safety equipment required for this work is not pictured. Figure 6 shows the pictures of the partially assembled experimental setup. The assembled equipment can supply up to 80 SLPM of NH₃ combined with 100 SLPM of air and 10 SLPM of propane fuel used to preheat the device at the startup. The liquid ammonia is kept at desired saturation pressure by resistive heaters placed around the circumference.



Figure 5: (A) Experimental setup used for reforming study (B) Cross-section of Swiss-roll reformer



Figure 6: Picture of the experimental setup. Arrows indicate the major flow path. The Swiss-roll device (*right*) is shown without ceramic insulation for a better overview.

Temperatures are recorded using K-type thermocouples in an Inconel sheath connected to National Instruments CDAQ equipment. Upon reaching a steady state, a sample is collected and analyzed using the SRI Instruments 8610C Gas Chromatography system configured for measuring H₂, N₂, O₂, and CO₂. Under nominal conditions, the O₂ and CO₂ are not detected, while the H₂:N₂ ratio and reactant supply flow rates allow the calculation of the unreacted NH₃ content in the exhaust. Knowledge of H2 and NH3 contents can be converted to the reforming efficiency

3. Results and Discussion

Modeling Work

The results from the 0D adiabatic PSR via CHEMKIN employing GRI-Mech III detailed chemistry are shown in Figure 7. The results suggest increased hydrogen favorability with increasing ammonia concentration, temperature, and corresponding residence time at such temperature. With an ammonia-rich mixture, the conversion rate becomes significant starting from 1300 K and eventually reaches maximum yield corresponding to equilibrium with increasing temperature, provided sufficient residence time. Otomo's and Song's detailed chemistry models also produce similar results except for the additional effect of ammonia decomposition, which becomes significant at impractically large residence time for Swiss-roll reformer. Within a reasonable high-temperature timescale for the flame inside the Swiss-roll combustor (tres ~ 1x10⁻³ sec - 1x10⁻¹ sec), the results imply high-temperature flame of approximately ~2000 K will have to be sustained to achieve significant hydrogen yield.



Figure 7: Perfectly-stirred reactor H-atom percentage in hydrogen form at outlet at varying temperature and residence time for (*Left*) $\Phi = 1.5$ and (*Right*) $\Phi = 5.0$ with GRI-Mech III

For Swiss-roll combustors, excess enthalpy $[EE \equiv (T_{max} - T_{inlet})/(T_{ad} - T_{inlet}) - 1]$ is a commonly-used design parameter, quantifying the amount of recirculated thermal enthalpy. The value can be increased by employing combustors with recirculation-favored designs (added turns, fins, etc.) at the expense of increased pressure drop. Aside from hydrogen yield favorability with increasing temperature, the yield also increases with the equivalence ratio Φ . However, it becomes increasingly challenging to operate with fuel-rich mixtures due to the temperature rise from combustion becoming progressively smaller. The effect of residence time is emphasized through Figure 8, where for shorter available residence time, the hydrogen yield always increases with increasing temperature up to 2500 K. With increased available residence time, the yield saturates at a specific temperature value, which decreases with increasing residence time.



Figure 8: Hydrogen yield at varying temperatures, residence time and required excess enthalpy values corresponding to different equivalence ratios.





Figure 9 above shows 2D simulation results for Swiss-roll combustors at a 1-ft scale operating with fuel-rich ammonia-air mixtures at 10 atm across high flowrate cases and two different Swiss-roll designs. The primary H species composition at combustor outlet mainly includes H_2O (not shown), H_2 and unprocessed NH₃. The amount of H_2O forms correspondingly to the amount of

Sub Topic: Novel Combustion Techniques

available O atoms (solely dependent on Φ). The H₂ yield dependency on Φ follows the expected trend following the 0D PSR results. The initial rise in H₂ yield going from stoichiometric to slightly fuel-rich ($1 \le \Phi \le 2$) is attributed to the composition effect since the temperature is sufficiently high in this region. The following drop in H₂ yield as fuel is increased further ($\Phi \ge 2$) is due to insufficient temperature, in which the temperature becomes the primary factor determining the yield. Most of the results shown are out-of-center flames, as the combination of flowrate and composition generally produces exceedingly flammable mixtures such that the flame does not require the full recirculation benefits from the combustor. Hence the flame retreats outward along the inlet channels. The yield favors higher flow rate cases (high Re) due to increasing temperature as overall enthalpy flux is increased, and the effect of heat loss becomes less significant as the flow rate increases. With the increased heat-recirculation provided by finned walls, the results produce improved extinction limit and yield compared to the plain-wall Swiss-roll device.



Figure 10: H-atom distributions between (*Left*) NH₃, (*Right*) H₂ at combustor outlet of 1-ft scale (*Top*) plain-wall, and (*Bottom*) finned Swiss-roll with double internal surface area, 3.5 turns Inconel-718 combustors (1.25 cm channel, 0.25 cm wall, 9.25 cm approximated core diameter) operating with NH₃-air mixture at 10 atm pressure with flame suppressed from inlet channels.

Figure 10 shows the results from a similar setup to prior results but with the reaction artificially suppressed from the inlet channels as a thought experiment. This represents solutions in which flashbacks are suppressed, and the flame is allowed to reap the full recirculation benefit from the Swiss-roll combustor, allowing the flame to reach even higher temperatures. The resulting H_2 yield behavior, in the same manner as free-flame cases shown in Figure 10, follows a bell-shaped curve with composition effect dominating for slightly-rich mixtures and temperature effect ruling the

ultra-rich mixtures. However, the yield instead favors low Re cases as the solution moves away from insufficient residence-time extinction limit characteristics to Swiss-roll combustors operating at a high flow rate. Additionally, for ultra-rich mixture cases ($4.5 \le \Phi \le 6$) for finned Swiss-roll, the maximum gas temperature is practically the same (~2000 K for $\Phi \approx 4.5$, ~1860K for $\Phi \approx 6.0$) across different Re values at the same Φ . This shows the secondary effect of high-temperature residence time, favoring lower flowrate cases provided sufficiently high temperatures.

Experimental Work

The early atmospheric pressure experiments succeeded in stabilizing the reactions at two inlet conditions. The flow configuration and related temperature profiles across the] Swiss-roll are presented in Table 1. In agreement with numerical predictions, the flame containment within the core section proved challenging. For example, based on the high Swiss-roll inlet temperature at $\Phi = 2.8$, it is apparent that the flame front flashed back and established itself within the inlet channel. The more fuel-rich $\Phi = 3.7$ case shows temperatures increasing through the core, although the reaction front is stretched into the outlet channel.

Equivalence Ratio Φ	Channel Velocity [m/s]	Temperature [°C]				
		Swiss-roll inlet	Core inlet	Core center	Core outlet	Swiss-roll outlet
2.8	7.5	964	892	910	875	807
3.7	11.7	190	861	882	890	712

Table 1: Experimental data: steady-state temperature profiles

The GC data has been at $\Phi = 2.8$ (EE = 4.75, 0.18s residence time) measured 21.3% H₂ and 60.4% N₂ with only a trace CO₂ signal below the instrument accuracy level. The missing 18.3% can be accounted for by the unreacted NH₃ and H₂O vapor, both outside the capabilities of the installed GC column. The measured H₂ is below the equilibrium prediction (31.5% H_{2,equilib}), supporting the importance of reaction temperature and residence time identified by the numerical study. The experiments are ongoing, with more results to follow.

4. Conclusions

Mixing hydrogen-rich reformate with ammonia is studied as a combustion augmentation method for enabling ammonia utilization in gas turbines. A novel Swiss-roll combustor has been proposed to provide the required reformate by an ultra-rich partial oxidation process. The numerical study identified the importance of Swiss-roll's excess enthalpy (EE) parameter, reaction temperature T_{R} , and core residence time t_{res} on expected H₂ yield. The best results are predicted for EE >2, TR > 2000 K, and $t_{res} > 0.01$ s. Notably, very high EE values can degrade reforming efficiency for severely insufficient T_R or t_{res} cases. The 2D study of Swiss-roll geometry supports the non-dimensional PSR findings while identifying the new issue of flame holding in the reactor core. The modeling results are further supported by early experimental results in a 6" Swiss-roll device successfully achieving steady-state operation at high equivalence ratios. The amount of generated hydrogen is sufficient to augment the burning properties of ammonia for gas turbines. The performed work supports the efficacy of the Swiss-roll reformer concept as a fuel pre-cracker for gas turbine applications.

5. Acknowledgments

The authors would like to thank the Department of Energy for financial support under the contract number DE-SC0022938.

6. References

- [1] A. Valera-Medina, F. Amer-Hatem, A.K. Azad, I.C. Dedoussi, M. De Joannon, R.X. Fernandes, P. Glarborg, H. Hashemi, X. He, S. Mashruk, J. McGowan, C. Mounaim-Rouselle, A. Ortiz-Prado, A. Ortiz-Valera, I. Rossetti, B. Shu, M. Yehia, H. Xiao, M. Costa, Review on ammonia as a potential fuel: From synthesis to economics, Energy and Fuels. 35 (2021) 6964–7029. https://doi.org/10.1021/acs.energyfuels.0c03685.
- [2] Z. Cesaro, M. Ives, R. Nayak-Luke, M. Mason, R. Bañares-Alcántara, Ammonia to power: Forecasting the levelized cost of electricity from green ammonia in large-scale power plants, Appl. Energy. 282 (2021) 116009. https://doi.org/10.1016/j.apenergy.2020.116009.
- [3] A.A. Khateeb, T.F. Guiberti, G. Wang, W.R. Boyette, M. Younes, A. Jamal, W.L. Roberts, Stability limits and NO emissions of premixed swirl ammonia-air flames enriched with hydrogen or methane at elevated pressures, Int. J. Hydrogen Energy. 46 (2021) 11969–11981. https://doi.org/10.1016/j.ijhydene.2021.01.036.
- [4] K.D.K.A. Somarathne, S. Hatakeyama, A. Hayakawa, H. Kobayashi, Numerical study of a low emission gas turbine like combustor for turbulent ammonia/air premixed swirl flames with a secondary air injection at high pressure, Int. J. Hydrogen Energy. 42 (2017) 27388–27399. https://doi.org/10.1016/j.ijhydene.2017.09.089.
- [5] D. Pugh, A. Valera-Medina, P. Bowen, A. Giles, B. Goktepe, J. Runyon, S. Morris, S. Hewlett, R. Marsh, Emissions Performance of Staged Premixed and Diffusion Combustor Concepts for an NH3/Air Flame With and Without Reactant Humidification, J. Eng. Gas Turbines Power. 143 (2021). https://doi.org/10.1115/1.4049451.
- [6] J. Li, S. Lai, D. Chen, R. Wu, N. Kobayashi, L. Deng, H. Huang, A Review on Combustion Characteristics of Ammonia as a Carbon-Free Fuel, Front. Energy Res. 9 (2021). https://doi.org/10.3389/fenrg.2021.760356.
- [7] H. Kobayashi, A. Hayakawa, K.D.K.A. Somarathne, E.C. Okafor, Science and technology of ammonia combustion, Proc. Combust. Inst. 37 (2019) 109–133. https://doi.org/10.1016/j.proci.2018.09.029.
- [8] A. Valera-Medina, M. Gutesa, H. Xiao, D. Pugh, A. Giles, B. Goktepe, R. Marsh, P. Bowen, Premixed ammonia/hydrogen swirl combustion under rich fuel conditions for gas turbines operation, Int. J. Hydrogen Energy. 44 (2019) 8615–8626. https://doi.org/10.1016/j.ijhydene.2019.02.041.
- [9] M. Ditaranto, I. Saanum, J. Larfeldt, Experimental Study on High Pressure Combustion of Decomposed Ammonia: How Can Ammonia Be Best Used in a Gas Turbine?, in: Vol. 3B Combust. Fuels, Emiss., American Society of Mechanical Engineers, 2021: pp. 1–8. https://doi.org/10.1115/GT2021-60057.
- [10] R.B. Snyder, Alternative control techniques document. NOx emissions from stationary gas turbines, Cary, NC, 1993.
- [11] P. Glarborg, J.A. Miller, B. Ruscic, S.J. Klippenstein, Modeling nitrogen chemistry in combustion, Prog. Energy Combust. Sci. 67 (2018) 31–68. https://doi.org/10.1016/j.pecs.2018.01.002.
- [12] V. Hacker, K. Kordesch, Ammonia crackers, in: W. Vielstich, A. Lamm, H.A. Gasteiger (Eds.), Handb. Fuel Cells – Fundam. Technol. Appl., John Wiley & Sons, Ltd, 2003: pp. 121–127.
- S.A. LLOYD, F.J. WEINBERG, A burner for mixtures of very low heat content, Nature. 251 (1974) 47–49. https://doi.org/10.1038/251047a0.
- [14] A.R. Jones, S.A. Lloyd, F.J. Weinberg, Combustion in heat exchangers, Proc. R. Soc. London. A. Math. Phys. Sci. 360 (1978) 97–115. https://doi.org/10.1098/RSPA.1978.0059.
- [15] I. V. Novosselov, P.C. Malte, S. Yuan, R. Srinivasan, J.C.Y. Lee, Chemical reactor network application to emissions prediction for industial DLE gas turbine, Proc. ASME Turbo Expo. 1 (2006) 221–235. https://doi.org/10.1115/GT2006-90282.
- [16] G.P. Smith, D.M. Golden, M. Frenklach, N.W. Moriarty, B. Eiteneer, M. Goldenberg, C.T. Bowman, R.K. Hanson, S. Song, W.C.J. Gardiner, V. V. Lissianski, Z. Qin, GRI-Mech 3.0, (2023). http://www.me.berkeley.edu/gri_mech/.