



A highly efficient autothermal microchannel reactor for ammonia decomposition: Analysis of hydrogen production in transient and steady-state regimes



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HIGHLIGHTS

- A scale-up of an autothermal microchannel reactor design for H₂ production.
- Effective thermal coupling between NH₃ decomposition and NH₃ oxidation reactions.
- Reactor performance assessed in transient and steady-state operating regimes.
- Improved reactor thermal efficiency of 75.9% and H₂ production rate of 0.71 kW_e.
- Combustible fuel-oxygen equivalence ratio identified to yield high N₂ product yield.

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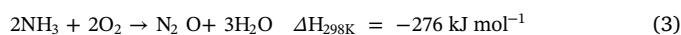
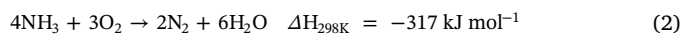
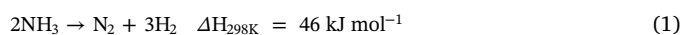
ABSTRACT

The experimental evaluation of an autothermal microchannel reactor for H₂ production from NH₃ decomposition is described. The reactor design incorporates an autothermal approach, with added NH₃ oxidation, for coupled heat supply to the endothermic decomposition reaction. An alternating catalytic plate arrangement is used to accomplish this thermal coupling in a cocurrent flow strategy. Detailed analysis of the transient operating regime associated with reactor start-up and steady-state results is presented. The effects of operating parameters on reactor performance are investigated, specifically, the NH₃ decomposition flow rate, NH₃ oxidation flow rate, and fuel-oxygen equivalence ratio. Overall, the reactor exhibits rapid response time during start-up; within 60 min, H₂ production is approximately 95% of steady-state values. The recommended operating point for steady-state H₂ production corresponds to an NH₃ decomposition flow rate of 6 NL min⁻¹, NH₃ oxidation flow rate of 4 NL min⁻¹, and fuel-oxygen equivalence ratio of 1.4. Under these flows, NH₃ conversion of 99.8% and H₂ equivalent fuel cell power output of 0.71 kW_e is achieved. The reactor shows good heat utilization with a thermal efficiency of 75.9%. An efficient autothermal reactor design is therefore demonstrated, which may be upscaled to a multi-kW H₂ production system for commercial implementation.

1. Introduction

Recently hydrogen (H₂) received extensive consideration as a clean energy source through fuel cell applications [1–4]. However, global capacity relating to H₂ storage and distribution infrastructure is not yet established; it remains one of the greatest factors impeding the development of a H₂ energy economy [5,6]. Hydrogen also supports a low volumetric energy density. Hydrogen dense energy carriers such as ammonia (NH₃), methanol (CH₃OH) and ethanol (C₂H₅OH) are thus suitable for transport and on-site H₂ generation [1–4,6–9]. On the other hand, a recent universal trend has shaped to distance from fossil fuels and carbon-containing compounds for power generation [10,11]. As a

carbon-free energy carrier, NH₃ is a particularly attractive option for H₂ generation as it supports a hydrogen density of 108.5 kg m⁻³ at 8.57 bar_a (saturated liquid at 20 °C) [12]. Also, NH₃ has a large global production capacity of 100 million tons y⁻¹ [5]. The process of NH₃ decomposition (often called reforming) produces H₂ through a slightly endothermic reaction (reaction 1). This reaction takes place over a suitable catalyst (Ru or Ni) at temperatures of 500–700 °C [13].



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Nomenclature

| | |
|-------------------|---|
| H | Height, m |
| ΔH_{298K} | Enthalpy of reaction, J mol ⁻¹ |
| L | Length, m |
| LHV_{H_2} | Hydrogen lower heating value, MJ kg ⁻¹ |
| LHV_{NH_3} | Ammonia lower heating value, MJ kg ⁻¹ |
| \dot{m}_i | Mass flow of component i, kg s ⁻¹ |
| \dot{n}_i | Mole flow of component i, mole s ⁻¹ |
| P_{eq} | Equivalent fuel cell power, W |
| W | Width, m |
| X_{NH_3} | Ammonia conversion, % |

Greek symbols

| | |
|----------|----------------------|
| η | Thermal efficiency |
| θ | Hydrogen utilization |

Subscripts & superscripts

| | |
|--------|---------------------|
| a | Absolute |
| decomp | Decomposition |
| e | Electric |
| in | Inlet |
| i | Mixture component i |
| out | Outlet |
| oxi | Oxidation |

Abbreviations

| | |
|-------|-------------------------------|
| db | Dry basis |
| GC | Gas chromatograph |
| SCR | Selective catalytic reduction |
| vol.% | Volume percent |
| wt.% | Weight percent |



Although NH₃ decomposition is a convenient process for on-site H₂ generation, the endothermicity of reaction makes it dependent on a heat source [14]. As such, self-sustained (autothermal) reactor operation is necessary for independent and continuous H₂ supply in off-grid locations [13,15]. Thermal coupling of chemical processes is well-known and a powerful tool to increase process efficiency and reduce equipment size and cost [16]. To simplify logistical issues, NH₃ oxidation is considered to supply in the thermal requirements of the decomposition reaction. Consequently, a single transported fuel is required for H₂ generation. Ammonia oxidation (reaction 2) is a viable heat source, as atmospheric nitrogen (N₂) and water are the main product species. Other possible oxidation products include nitrous oxide (N₂O) and nitric oxide (NO) through reactions 3–4, however not desired in this application.

Heat supply from NH₃ oxidation/combustion is sustainable and may take place homogeneously (flame) or catalytically. The simplicity of homogeneous combustion is an attractive prospect to satisfy thermal requirements. However, to achieve improved heat and mass transfer in small-to-medium-scale H₂ generation applications, homogeneous combustion may be susceptible to thermal and radical quenching [2,17,18]. In contrast, catalytic combustion supports broader operating limits and has the ability to improve fuel conversion at much lower reaction temperatures [16,18,19]. Material and catalyst stability is therefore

sustainable at realistic flow velocities. In previous studies, lower reactor temperatures and using N₂ selective catalysts (e.g. Pt/Al₂O₃) proved to significantly reduce N₂O and NO_x formation [20,21]. Furthermore, fuel rich conditions influence thermodynamic equilibrium in favor of N₂ formation through so-called selective catalytic reduction (SCR) reactions [10,11,20].

To achieve distributed H₂ generation through NH₃ reforming, compact reactor setups are necessary. Also, an efficient thermal coupling between the endothermic NH₃ decomposition and exothermic NH₃ oxidation reaction is pre-requisite [2,19]. Microchannel reactor technology is easily scaled to desired generation capacity, and proves convenient when incorporated in modularized plants [14,18,19]. These reactors have characteristic small diffusion paths and essentially support fast reaction rates through process intensification [16,22]. A central heat-conducting plate is characteristic between spatially coupled microchannels [19]. A wide range of process conditions, reactions, catalysts and flow configuration (co-, counter- and cross-flow) contribute to the multi-functionality of these reactors [23]. The use of cocurrent flow configurations proved beneficial in the coupling with a highly exothermic reaction [17,24]. Enhanced heat transfer was achieved at the reactor inlet region where high reaction rates dominate. Similar thermal behavior is envisioned in the coupling with the highly exothermic NH₃ oxidation reaction.

Autothermal NH₃ decomposition in a microchannel reactor was the

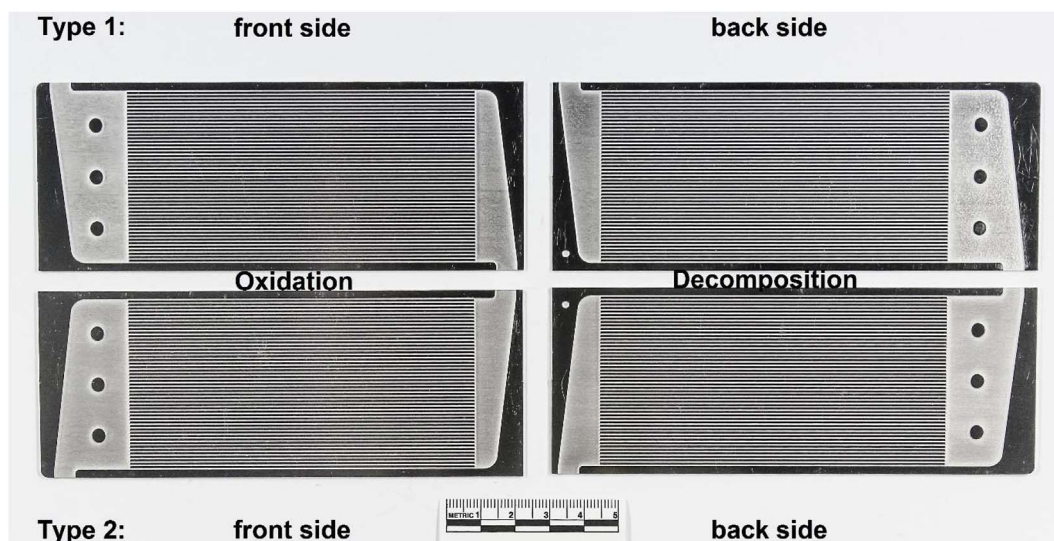


Fig. 1. Alternating microchannel plate arrangement of NH₃ oxidation (left) and NH₃ decomposition (right) reaction zones.

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