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Thermodynamic Analysis of Catalytic Urea Steam Reforming for Decentralized Hydrogen Production

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The increasing interest in integrating hydrogen (H₂) into the global energy mix, alongside the challenges associated with its storage and transportation, has motivated the development of novel technologies for decentralized H2 production. In this context, hydrogen carriers are promising candidates to generate H2 on-site and on-demand through reforming reactions. When these reformers are coupled with fuel cells (FCs), H2 can be efficiently converted into electricity, making this approach attractive for transport (onboard) applications. Urea (CO(NH₂)₂) is a hydrogen vector that has been underexplored in the literature but holds potential due to its well-established supply chain, non-toxic nature, and affordability. Bearing this in mind, this work investigated urea steam reforming through a simulation study, using Aspen Plus software. Here, SR-Polar equation-of-state and the Gibbs reactor model were used to assess the H₂ production. Through sensitivity analyses, the effect of key parameters such as the steam-to-carbon (S:C) ratio, temperature, and pressure were systematically evaluated to identify the optimal operating conditions. It was found that at a S:C ratio of 5 and 0.5 bar, the reactor operating temperature could be reduced to 600 °C to produce 2.78 moles of H₂ per mole of urea, approaching the reaction stoichiometry of 3 molH₂/molurea. After process optimization, hydrogen purification was simulated to meet ISO 14687:2019 standards for application in a Proton-exchange membrane fuel cell (PEMFC), and the power output was calculated for this type of FC, focusing on the potential of the system in decentralized hydrogen generation.

* 1. Introduction

The demand for hydrogen (H2) is increasing worldwide due to its potential to contribute to the reduction of greenhouse gases (GHG) emission in sectors that are challenging to decarbonize, such as heavy industry and transportation, where viable alternatives are limited or difficult to adopt (IEA, 2023). Additionally, a report from the Hydrogen Council states that a 60% reduction in global carbon emissions by 2050 is imperative to mitigate climate change (Hydrogen Council, 2017). However, according to the International Energy Agency (IEA), the current global hydrogen production comes mainly from fossil fuels, with only less than 1% being originated from renewable sources (IEA, 2023). Therefore, considering today’s energy transition context and acknowledging the difficulties revolving around hydrogen storage and transportation, the utilization of hydrogen carriers as a hydrogen vector is introduced. These carriers are compounds that possess hydrogen in their molecule, such as methane, ethanol, ammonia, and urea, being presented as promising candidates to generate H2 on-site and on-demand through reforming reactions (Barros et al., 2024).

Among the available carriers that can be explored for H2 generation, urea (CO(NH₂)₂) is highlighted for being a non-toxic, non-flammable, and odorless crystalline substance (Rollinson et al., 2011a), which is considered an advantage compared to other hydrogen vectors in terms of safety concerns. Additionally, urea has a gravimetric hydrogen content of approximately 6.71 wt%, it can be produced synthetically on a commercial scale and is also present in nature through animal excretions and plants as nitrogen storage (Rollinson et al., 2010). This, combined with the fact that urea is biodegradable, makes it a very interesting hydrogen carrier and object of study. However, only a few works report urea utilization for H2 production (Dupont et al., 2013; Rollinson et al., 2010; Rollinson et al., 2011b). In this sense, the present study uses Aspen Plus software to explore the urea steam reforming (USR) for decentralized hydrogen generation, aiming at fuel cells (FCs) application. Although USR simulation has been previously reported in the literature (Dupont et al., 2013), to the best of our knowledge, this is the first work simulating USR in Aspen Plus and analysed for the proposed utilization.

While the catalytic reforming of urea can yield 2 moles of H₂ per mole of CO(NH₂)₂, steam reforming offers the advantage of generating 3 moles of H₂ per mole of urea (3 molH₂/molurea). Therefore, to enhance hydrogen production, sensitivity analyses were conducted to investigate the influence of key operating parameters, including the steam-to-carbon (S:C) molar ratio, temperature, and pressure, on hydrogen yield. The results indicated that increasing the S:C molar ratio and operating at a reduced pressure of 0.5 bar enabled a higher hydrogen yield at a relatively moderate temperature of 600 °C. After determining the operating conditions, hydrogen purification was carried out in a membrane, respecting the standards determined by ISO 14687:2019 required for a Proton-exchange membrane fuel cell (PEMFC) (Floriam et al., 2024). To meet these standards, initial calculations were performed in Excel and subsequently implemented in the separator model (Sep) within the simulation software. Finally, the power output equivalent (Peq) for the PEMFC was calculated based on the hydrogen production rate (in kg/s), using an equation from the literature (Engelbrecht et al., 2018).

* 1. Methodology
		1. Process Description and Simulation Approaches

This study was carried out at Aspen Plus® V10 software and focused on urea steam reforming for decentralized hydrogen production. Here, the effect of S:C ratio (kilomoles of water per kilomole of urea), temperature, and pressure on hydrogen production were evaluated. The process configuration, shown in Figure 1, was simulated under steady-state conditions, using SR-POLAR as the thermodynamic method, which is based on the Schwartzentruber-Renon equation-of-state. The method is suitable for nonpolar and strongly polar compounds as well as highly nonideal systems (AspenTech, 2001). All components, except for urea (a solid in its natural state), were considered conventional for the selection of input compounds in the software.

Initially, two streams—one comprising pure urea (designated as stream UREA) and the other consisting of pure water (designated as stream WATER)—were combined in a mixer (MX1) prior to being introduced into the reforming reactor (REFORMER). The *RGibbs* model was selected as the reformer as it predicts the composition of the product at both in phase and chemical equilibria by minimizing the Gibbs free energy (Barros, 2024). After this step, the hydrogen purification was performed in a separator *Sep* model (MEMBRANE) to obtain hydrogen purity that meets the standards of ISO 14687:2019 for a PEMFC. The contamination limits of the main components found in purified hydrogen are specified in Table 1, and the recovery required to achieve these limits was calculated using Microsoft Excel. The calculated values were input into the MEMBRANE block to ensure effective separation of the components. Finally, given that the purified hydrogen is directed to a PEMFC for electricity generation, the equivalent FC power output, Peq, was calculated in Watts for this process.



Figure 1: Description of the urea steam reforming process for hydrogen production.

Table 1: Specifications of ISO 14687:2019 for Hydrogen fuel quality

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| --- | --- | --- |
| H2 Fuel Index  | Total non-hydrogen gases | Max. Conc. Individual Contaminants |
| 0.9997 | 300 µmol/mol | Water (H2O) | 5 µmol/mol |
|  | Methane (CH4) | 100 µmol/mol |
| Oxygen (O2) | 5 µmol/mol |
| Nitrogen (N2) | 300 µmol/mol |
| Carbon dioxide (CO2) | 2 µmol/mol |
| Carbon monoxide (CO) | 0.2 µmol/mol |
| Ammonia (NH3) | 0.1 µmol/mol |

* + 1. Equations Involved in the Reforming Reaction

The main overall reaction involved in USR is given in Eq(1), which depicts the production of 3 mol of H2 for a stoichiometric feed of 1 molH2O:1 molurea. At the same time, Eq(2) shows the overall reaction of the catalytic thermal decomposition of urea (in the absence of water), producing only 2 moles of H2 per mole of urea consumed. In addition to these equations, other parallel reactions can also occur during reactor operation, such as the methanations of CO and CO2 (Eq(3) and Eq(4), respectively), the water gas shift reaction (WGSR) in Eq(5), the ammonia (NH3) synthesis in Eq(6), and the urea cracking into isocyanic acid (HCNO) and ammonia in Eq(7). The production of solid carbon (coke) is also possible during reforming reactions, but it is outside the scope of this work and will not be presented in the results section. All compounds involved in the reactions presented from Eq(1) to Eq(7) were included in the simulation input to evaluate their potential formation. Additionally, nitrogen oxides (NOx), specifically NO and NO₂, were added to the simulation input to assess their possible generation through the reverse selective non-catalytic oxidation reaction as shown in Eq(8).

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| $CO\left(NH\_{2}\right)\_{2}+H\_{2}O\rightarrow 3H\_{2}+CO\_{2}+N\_{2}$ $∆H\_{25 ºC}^{0}=80.4 kJ/mol$ | (1) |
| $CO(NH\_{2})\_{2}\rightarrow 2H\_{2}+CO+N\_{2}$ $∆H\_{25 ºC}^{0}=125 kJ/mol$ | (2) |
| $CO+3H\_{2}\leftrightarrow CH\_{4}+H\_{2}O$ $∆H\_{25 ºC}^{0}=-206.2 kJ/mol$ | (3) |
| $CO\_{2}+4H\_{2}\leftrightarrow CH\_{4}+2H\_{2}O$ $∆H\_{25 ºC}^{0}=-165 kJ/mol$ | (4) |
| $CO+H\_{2}O\leftrightarrow H\_{2}+CO\_{2}$ $∆H\_{25 ºC}^{0}=-41.2 kJ/mol$ | (5) |
| $N\_{2}+3H\_{2}\leftrightarrow 2NH\_{3}$ $∆H\_{25 ºC}^{0}=-91.9 kJ/mol$ | (6) |
| $CO(NH\_{2})\_{2}\rightarrow HCNO+NH\_{3}$ $∆H\_{25 ºC}^{0}=87.9 kJ/mol$ | (7) |
| $2N\_{2}+3H\_{2}O\leftrightarrow 2NH\_{3}+NO\_{2}+NO$ $∆H\_{25 ºC}^{0}=757.1 kJ/mol$ | (8) |

* + 1. Sensitivity Analyses

In order to enhance hydrogen production, sensitivity analyses were carried out by simultaneously varying two parameters. In the first analysis, the S:C molar ratio was evaluated in the range of 1 to 10, together with the temperature that was tested between 200 ºC to 1000 ºC, with an increment of 20 ºC. In this case, the reactor pressure was fixed at 1 bar. Based on the first analysis and the authors’ interpretation, the S:C ratio was defined and fixed for the second sensitivity analysis, which assessed the reactor pressure in the range from 0.5 to 2 bar, with 0.5 bar steps, and temperature from 200 ºC to 1000 ºC, with 20 ºC steps. Based on these evaluations, the operating parameters were defined to enable the calculation of the reaction products and the hydrogen yield ($Y\_{H\_{2}}$), as presented in Eq(9). Here, $F\_{H\_{2,out}}$ is the molar flow rate of the outlet hydrogen, $F\_{fuel, in}$ is the inlet molar flow rate of urea, and $v\_{H\_{2}}$ is the theoretical stoichiometric coefficient of the produced H2 (Eq(1)) (Di Nardo et al., 2024). After these steps, hydrogen purification was simulated and the power output was calculated.

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| --- | --- |
| $Y\_{H\_{2}}=\frac{F\_{H\_{2,out}}}{F\_{urea, in}\*v\_{H\_{2}}}\*100\%$  | (9) |

* + 1. Hydrogen Purification and Power Output Equivalent

As previously explained, the purification process was carried out using a *Sep* model separator, which was considered to represent a membrane positioned at the outlet of the reformer. The separation fraction of each component was filled in the simulator based on results obtained in Excel, which were calculated respecting ISO 14687:2019 standards as indicated in Table 1 for PEMFCs. After that, the hydrogen production calculated in the software, based on the operating conditions defined by the sensitivity analyses, was converted into kg/s to calculate the power generated for application in a PEM fuel cell. The calculation of the power output, Peq (kW), is presented in Eq(10). Here, $η\_{PEM}$ is the energy efficiency (%) for a PEMFC and $θ\_{PEM}$ is the hydrogen utilization (%) for a PEMFC, both assumed from literature as 60% and 80%, respectively, while $\dot{m}\_{H\_{2}}$ is the mass production rate of hydrogen (kg/s), and $LHV\_{H\_{2}}$is the lower heating value of hydrogen (120 MJ/kg) (Engelbrecht et al., 2018).

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| $P\_{eq}=η\_{PEM}θ\_{PEM}\dot{m}\_{H\_{2}}LHV\_{H\_{2}}$  | (10) |

* 1. Results

Figures 2a and 3a illustrate the graphical results derived from the sensitivity analyses. The first graph, Figure 2a, shows the effect of simultaneously varying the S:C ratio from 1 to 10 and temperature from 200 to 1000 ºC on hydrogen production. Here, the pressure is fixed at 1 bar. As can be observed, for an equimolar feed of urea and water (1 kmol of H2O to 1 kmol of CO(NH₂)₂), the maximum H2 production is achieved at a temperature of 760 ºC, being limited to only ~2.26 kmol of H2 per kmol of urea, representing a yield of less than 75.50%. By increasing the S:C ratio, a remarkable improvement in H2 production is obtained, with a maximum H2 yield of ~96.20% at a S:C ratio of 10 and 580 ºC, representing a production of more than 2.88 kmolH2/kmolurea. This phenomenon was likewise reported by Dupont et al. (2013), who attributed the increase in hydrogen yield at higher steam-to-carbon (S:C) ratios to Le Chatelier's Principle, where the additional H₂O shifted the equilibrium, promoting greater consumption of reactants and consequently enhancing hydrogen generation.

Interestingly, increasing the S:C molar ratio also causes a shift in the temperature at which H₂ production is maximized, with lower temperatures being reached as the S:C ratio increases. Although the main overall reaction in USR is endothermic, several parallel reactions occur during the reforming process (see Eq(1) to Eq(8)). When the water content in the system increases, the WGSR, presented in Eq(5), is favoured. Since this reaction is exothermic, the equilibrium shifts toward lower temperatures, which explains the behaviour observed in Figure 2a. This statement can be reinforced by Figure 2b, which shows the production of CO2 (in blue) and CO (in red) as a function of temperature and S:C ratio (ratios of 1, 5 and 10, represented by the inset symbols in the figure). According to the graph, the production of CO at temperatures up to 500 °C is close to zero for all the evaluated ratios. This is assigned to the fact that at lower temperatures, the exothermic reactions of methanation of CO (Eq(3)) and WGS (Eq(5)) are favoured, leading to the formation of CH4 and CO2, respectively. This phenomenon is further evidenced by the increase in CO2 production in this graph region, particularly for elevated S:C ratios. This is attributed to the displacement of the WGS equilibrium, where the excess water is consumed in conjunction with CO, to form H2 and CO2. At the same time, in the case of very high temperatures, since the WGS and methanation reactions are exothermic, the equilibrium tends to move to the left, towards the reactants. Therefore, in high temperatures, CO production increases significantly while CO2 generation decreases, especially for the S:C ratio of 1. Even so, compared to the CO produced, the amount of CO2 at the product stream remains relatively high for the S:C ratios of 5 and 10, since the methanation reaction is very exothermic, so the inverse direction of Eq(4) can also lead to the generation of CO2.

Figure 2: a. Hydrogen production as a function of S:C ratio and temperature, determined at a fixed pressure of 1 bar by sensitivity analysis. b. CO and CO2 production as a function of S:C ratio and temperature.

Although cost assessment is out of the scope of this study, increasing the water proportion in the system requires higher energy input to generate steam, which significantly raises the overall process costs. In light of this, the authors opted to employ an intermediate S:C ratio of 5, as it can effectively improve hydrogen yield to values exceeding 90% at temperatures between 600 and 720 ºC. After defining the S:C ratio, a second sensitivity analysis was conducted to evaluate the combined effects of pressure and temperature on H2 yield, as shown in Figure 3a. The results indicate that increasing system pressure reduces hydrogen production, while operating under a little vacuum facilitates reactant conversion in the reforming reaction and lowers the temperature at which maximum H2 generation occurs. These observations can also be explained by Le Chatelier's Principle, which predicts that reducing reactor pressure shifts the equilibrium toward the gaseous reaction with the greatest number of moles. In this case, the reforming reaction benefits, resulting in higher H₂ production at reduced pressures (Ismaila et al., 2021). At atmospheric pressure and an S:C ratio of 5, the maximum hydrogen yield is approximately 91.50% (2.74 kmolH₂/h) at 640 ºC. In contrast, at 0.50 bar and the same S:C ratio, the maximum yield increases slightly to ~92.70% (2.78 kmolH₂/h), achieved at a lower temperature of 600 ºC. While the reduction in system pressure results in only a modest improvement in H₂ production, it leads to a significant decrease in reactor operating temperature. Additionally, the temperature range for achieving hydrogen yields above 90% expands to be between 540 and 720 ºC.

Figure 3: a. H2 production as a function of pressure and temperature, determined at a fixed S:C ratio of 5 by sensitivity analysis. b. Products generated as a function of temperature, at fixed S:C ratio of 5 and 0.5 bar.

To quantify the products generated during the USR process, Figure 3b was constructed, illustrating the principal reaction products under operating conditions with a S:C ratio of 5, pressure of 0.5 bar, and temperatures ranging from 200 to 1000 ºC. According to the graph, H2 is the primary product formed at temperatures above 340 ºC, being maximized at 600 ºC. Additionally, due to the exothermic nature of the methanation reactions, Eq(3) and Eq(4), CH4 production approaches zero at temperatures above 520 ºC, shifting the reactions toward the reverse direction, favouring the generation of CO and CO₂. The production of NH₃ (Eq(6)) and HCNO (Eq (7)) is not shown, as the amount of these compounds were found to be negligible. Furthermore, the analysis of NOₓ in the product stream indicated that these pollutants were not generated during the process, as expected, since it is an extremely endothermic and thermodynamically unfavourable reaction according to Eq(8). Therefore, one advantage of the USR is its relatively low GHG emissions. However, a comprehensive analysis of the entire process life cycle —such as a life cycle assessment (LCA), encompassing urea production, reforming reaction, and hydrogen application in fuel cells— is necessary to quantify the total GHG emissions.

Based on the findings presented above, the selected operating conditions for the reformer were a S:C ratio of 5, a pressure of 0.5 bar, and a temperature of 600 ºC. The hydrogen purification step was subsequently simulated, assuming that the membrane separation process meets ISO standards, as outlined in Table 1. To achieve this, the membrane would need to exhibit high selectivity for hydrogen and block the contaminants. Additionally, other critical properties of the membrane, such as chemical, thermal, and mechanical stability, should be considered for practical usage (Du et al., 2021). Based on the selected operating conditions for the reformer, the separation fractions of the various components were calculated using Excel. The results, detailing the separation fractions of compounds that permeate the membrane towards the PEMFC, are summarized in Table 2. As can be seen, a negligible fraction of contaminants would be tolerated in the PEMFC, therefore an advanced separation membrane would be required in a practical system to allow such a level of separation.

Table 2: Split Fraction of components permeating the membrane for utilization in the PEMFC.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| H2  | H2O | CO | CO2 | CH4 | NH3 | N2 |
| 1 | 3.30 x 10–6  | 2.85 x 10–6 | 6.96 x 10–6 | 4.79 x 10–2 | 7.09 x 10–4 | 5.36 x 10–4 |

Once properly purified, the produced hydrogen could be applied to PEM fuel cells for power generation. The total H2 produced for the reformer operating conditions (S:C ratio of 5, 600 ºC and 0.5 bar) was 2.78 kmolH2/h, which corresponds to a mass production rate of ~1.56 x 10–3 kg/s (5.60 kg/h). Therefore, by using Eq(10) to calculate Peq, the power output for the proposed system corresponds to 89.67 kW. Comparing the proposed USR system with the Toyota Mirai commercial car, which is powered by hydrogen for PEMFCs and operates at a maximum power of 128 kW, it would be necessary to increase the reactants feed of the USR system to reach a 42% higher power output. Therefore, further improvements would be required for onboard and non-centralised hydrogen production from urea.

* 1. Conclusions

This study proposed a decentralized approach to hydrogen production via urea steam reforming. The thermodynamic evaluation of the process was conducted using Aspen Plus, with sensitivity analyses performed to optimize the reformer operating parameters, including the S:C ratio, pressure, and temperature. Based on these analyses, the optimal conditions were identified as a S:C ratio of 5, a temperature of 600 °C, and a pressure of 0.5 bar. Subsequently, calculations for hydrogen purification were performed to comply with ISO 14687:2019 standards, and a corresponding power output of 89.67 kW was estimated. Despite the significant power output, the feed flow rate of this system would need to be increased, for instance, to approach the performance of the Toyota Mirai commercial vehicle, which produces a maximum power of 128 kW. Furthermore, improvements in the process are essential, as the proposed system would require substantial amounts of urea and water to produce the necessary H2 for hydrogen-powered vehicle applications.

Nomenclature

$F\_{H\_{2,out}}$ – molar flow rate of outlet hydrogen, kmol/h

$F\_{urea, in}$ – molar flow rate of inlet urea, kmol/h

$LHV\_{H\_{2}}$ – lower heating value of hydrogen, MJ/h

$\dot{m}\_{H\_{2}}$ – mass production rate of hydrogen, kg/h

$v\_{H\_{2}}$ – Theoretical stoichiometric coefficient of H2

$P\_{eq}$ – power output equivalent, We

$θ\_{PEM}$ – hydrogen utilization for a PEMFC, %

$η\_{PEM}$ – Energy efficiency for a PEMFC, %

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