Hydrous Hydrazine decomposition over Rh/Al_2O_3 catalyst: CFD studies

Panayiota Adamou[1], Silvio Bellomi[2], Eleana Harkou[1], Alberto Villa[2], Nikolaos Dimitratos[3,4], Achilleas Constantinou[1]*

1 Department of Chemical Engineering Cyprus University of Technology, 57 Corner of Athinon and Anexartisias, 3036 Limassol, Cyprus; 2 Dipartimento di Chimica, Universitá degli Studi di Milano, via Golgi, 20133 Milan, Italy; 3 Department of Industrial Chemistry "Toso Montanari", Alma Mater Studiorum University of Bologna, Viale Risorgimento 4, 40136 Bologna, Italy; 4 Center for Chemical Catalysis-C3, Alma Mater Studiorum University of Bologna, Viale Risorgimento 4, 40136 Bologna, Italy

**Corresponding author: a.konstantinou@cut.ac.cy*

Highlights

- H_2 generation from hydrous hydrazine decomposition over 0.5 wt.% Rh/Al_2O_3 commercial catalyst.
- Optimized stirring rate at 1400 rpm, accurately validated by CFD studies.
- Velocity profile is uniform in the batch system according to CFD models.

1. Introduction

Hydrogen (H_2) is a promising green alternative to fossil fuels that can be generated via various methods. Hydrous hydrazine ($N_2H_4.H_2O$), can be decomposed via two pathways, desirable dehydrogenation giving H_2, and less favorable ammonia (NH_3) formation [1]. The addition of alkali media is crucial to favor H_2 production over NH_3 and thus, batch reactors are usually utilized, due to the corrosive nature of the basic medium in continuous flow systems [2]. Computational Fluid Dynamics (CFD) can be used to validate experimental data and predict results by varying experimental parameters saving time and materials [3], [4]. This research aims to validate experimental data regarding H_2 generation from $N_2H_4.H_2O$ on a 0.5 wt% Rh catalyst supported on alumina (Al_2O_3). CFD studies are also conducted for the investigation of the uniformity of the system. Overall, this work contributes insights into hydrazine catalysis and reactor system considerations for sustainable H_2 production.

2. Modelling Methodology

A CFD model was developed to validate the experimental results on hydrogen selectivity varying different parameters. Since there is no inflow or outflow in a batch reactor and a perfectly mixed volume is assumed, a 0D model was developed using the COMSOL software. The mass balance for a component, i, is:

$$\frac{dc_i}{dt} = R_i \tag{1}$$

where c_i is the molar concentration of each species (mol/m^3), t is the time (s) and R_i expresses the sum of each components rate expressions (mol/m^3.s).

For the uniformity of the system, a 2D model was developed using the Rotating Machinery Turbulent Flow, k-ε. The continuity equation for conservation of mass (Eq.2) and Navier-Stokes equation for conservation of momentum (Eq.3) were used as shown below:

$$\rho \nabla \cdot u = 0 \tag{2}$$

where ρ is the density of the fluid (kg/m^3) and u is the velocity (m/s), for an incompressible fluid.

$$\rho \frac{\partial u}{\partial t} + \rho(u \cdot \nabla) = \nabla[-pI + \kappa] + F \tag{3}$$

where, on the right side of the equation, p is the pressure (Pa), I denotes the identity tensor, κ (Pa) is the viscous stress tensor and F expresses the external force (N/m^3).

3. Selected Results and discussion

Catalytic tests were conducted varying the stirring rate from 250 to 1400 rpm, to investigate external mass transfer limitations. As seen from Figure 1a stirring rate doesn't have a significant effect on the reaction and thus, mass transfer resistance is avoided. CFD studies exhibit great validation with the experimental data with H_2 selectivity over 92%. To ensure the achievement of kinetic regimes, 1400 rpm was used as the optimum stirring rate. To confirm that the batch system is uniform at 1400 rpm, 2D simulations were performed investigating velocity magnitude. Figure 1b shows that at 0.75s, influenced by the rotation of the magnetic stirrer, uniformity is achieved, with the only exception being the tips of the magnetic stirrer where velocity magnitude is higher. More parametric studies were conducted on temperature uniformity, distribution of the reactant and catalytic particles in the reactor but the results are not shown here.

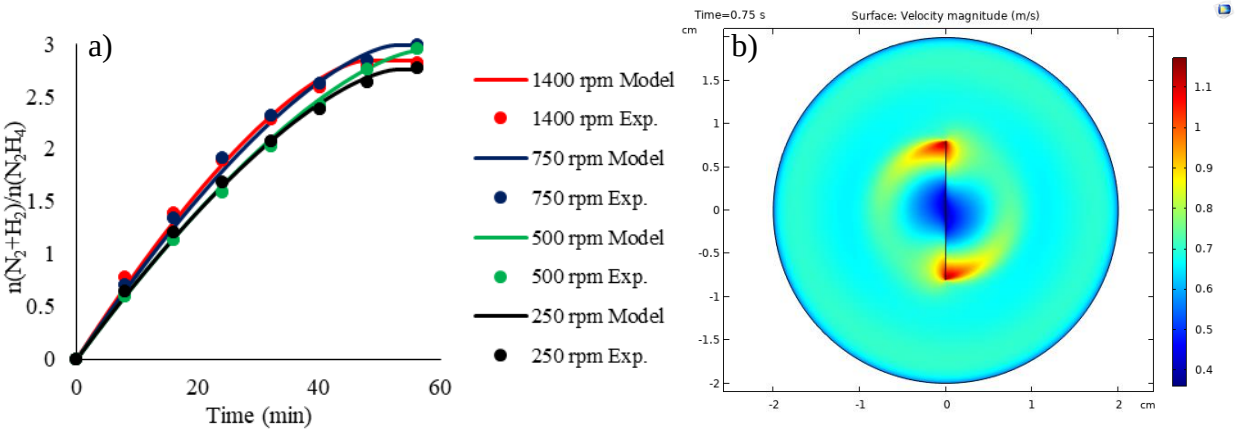

Figure 1. a) $n(H_2 + N_2)/n(N_2H_4)$ versus time varying the value of stirring rate at 250, 500, 750 and 1400 rpm. Reaction conditions: 150 µL of 3.3 M N_2H_4, 0.5 M NaOH, 1000:1 substrate to metal molar ratio and temperature of 70 ºC, **b)** Velocity field distribution in the batch system at 0.75s.

4. Conclusions

To conclude, this study validated experimental data on the catalytic decomposition of hydrous hydrazine on Rh/Al_2O_3 catalyst for H_2 generation using CFD studies. CFD simulations also provided a comprehensive understanding of the system's uniformity focusing on velocity profiles. According to the computational findings the system was uniform and thus agreeing with the experimental findings of high H_2 selectivity.

References

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Keywords

"hydrous hydrazine; hydrogen, batch, CFD".