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Trickle-bed reactors design for hydrogen chemical storage

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In recent years, molecular hydrogen has emerged as a promising future energy vector. However, its widespread adoption faces challenges related to sustainable production and safe, efficient storage. Chemical storage involves bonding hydrogen to other compounds, enabling long-term storage without high pressures or extremely low temperatures. Despite these advantages, current chemical storage methods often use unstable and highly reactive solid hydrides, posing significant safety risks despite their high storage density. To address these issues, this work focuses on the chemical hydrogen storage strategy based on the hydrogenation of benzene to cyclohexane. This method stores three moles of hydrogen per mole of cyclohexane, with hydrogen readily recoverable through the reverse dehydrogenation process. A critical step in implementing this system is designing an efficient reactor for benzene-to-cyclohexane conversion. A trickle bed reactor is identified as a viable candidate. However, the fluid dynamics complexity and the presence of flammable, reactive species necessitate detailed analysis to ensure an optimal, safe configuration. This study presents a comprehensive design protocol and modeling approach for the trickle bed reactor, addressing fluid dynamics, active bed length, and key mechanical design parameters. The proposed model provides a foundation for advancing safer chemical hydrogen storage solutions. To gain a broader understanding of the proposed solution, an economic analysis was also conducted to complement the technical assessment.

* 1. Introduction

The Net Zero Emissions by 2050 Scenario (NZE) anticipates global climate neutrality by 2050, with net-zero greenhouse gas (GHG) emissions (Carretta et al., 2024). Achieving this goal requires significant advancements in carbon capture technologies and the adoption of hydrogen as a key energy vector (Maporti et al., 2024). However, large-scale hydrogen production presents notable challenges, particularly in medium- and long-term storage. Current solutions, such as compressed hydrogen at pressures up to 1000 bar (Carboni et al., 2022) and cryogenic storage (Vianello et al., 2020), face limitations related to safety concerns and the high refrigeration costs needed to maintain hydrogen's extremely low boiling point (Sakamoto et al., 2021) and manage ortho-para hydrogen transitions (Riaz et al., 2023).

Chemical hydrogen storage offers an alternative approach, bonding hydrogen to stable compounds and eliminating the need for cryogenic cycles or high-pressure systems. Hydrogenation/dehydrogenation cycles (Pio et al., 2022) involving stable liquid compounds, such as benzene and cyclohexane (Andriani et al., 2024a), present an efficient and safer solution. Liquid-phase reactions provide higher storage densities and operational simplicity compared to solid hydride carriers (Wang et al., 2012). Benzene hydrogenation, which stores three moles of hydrogen per mole of benzene, is particularly promising but strongly exothermic, necessitating robust thermal management to prevent runaway reactions (Andriani et al., 2024b). A trickle-bed reactor is a suitable configuration for continuous liquid-phase benzene hydrogenation, offering scalable options for efficient heat removal and high productivity. Despite its advantages, comprehensive models for industrial-scale reactor design and performance analysis remain underdeveloped. This study proposes a detailed trickle-bed reactor model that addresses both functional and structural design aspects and economic evaluations, validated through a case study of the benzene/cyclohexane system as a safer hydrogen storage solution.

* 1. Methodology

The proposed methodology consists of 6 main steps: input settings (Section 2.1), fluid dynamic regime selection (Section 2.2), structural design (Section 2.3), functional design (Section 2.4), verification step (Section 2.5) and economic analysis (Section 2.6). These steps form an interconnected framework where initial assumptions are iteratively refined to meet design constraints. Figure 1 illustrates the workflow.



Figure 1. Logical workflow of the design procedure.

* + 1. Inputs settings

The first step defines key process parameters to establish the reactor configuration and address degrees of freedom. Trickle-bed reactors can adopt either a column or multitubular setup. The packed column configuration handles multiphase flow and can include a jacket for enhanced heat removal. Multitubular setups, by contrast, offer a higher surface area-to-volume ratio, making them preferable for highly exothermic reactions due to their superior heat transfer capabilities. Key parameters include inlet temperature, pressure, mass flow rates, reactor geometry, catalyst type, and kinetic and transport models. The inlet conditions must balance safety and performance, avoiding runaway reactions while ensuring adequate reactant conversion and product yield. Configuration choice depends on thermal needs: columns are cost-effective for processes with lower heat generation, while multitubular designs are better suited for reactions requiring significant heat dissipation. Catalyst selection and kinetic models are critical and may be derived from literature or experimental studies. Transport and thermodynamic models, such as those proposed by Ranade et al. (Ranade et al., 2011) and (Green and Southard, 2019), provide essential parameters for robust reactor modeling.

* + 1. Fluid dynamic regime

The second step involves selecting the appropriate fluid dynamic regime to ensure optimal multiphase flow. The co-current downflow configuration is favored for trickle-bed reactors due to its operational stability (Ranade et al., 2011) (Saroha and Khera, 2006). The main flow regimes include bubbling, trickle, pulse, and spray. Pulse flow offers high performance but is usually feasible only for small-to-medium-scale systems. For industrial-scale reactors, a trickle flow or a transition to pulse flow is more realistic. Flow regime selection relies on dimensionless numbers (λ, ξ, and ψ), which are functions of mixture properties such as density, viscosity, and surface tension. Evaluations are performed under standard conditions (1 bar, 298 K) (Rouhani and Sohal, 1983). These values map to coordinates on a flow regime chart, enabling calculations of mass flow rates per unit free surface area of the channel for gas and liquid. The overall gas and liquid ($L$) flow rates are then derived based on the channel diameter.

* + 1. Structural design

The structural design phase calculates component thicknesses to withstand internal stresses, following ASME Boiler and Pressure Vessel Code (2023) guidelines. Assumptions include no corrosion allowance and zero wall undertolerance for simplicity. Reactor head thicknesses are conservatively set equal to the cylindrical sections due to the lower pressure confinement capacity of non-cylindrical shapes. Structural integrity is critical for maintaining operational safety and supporting the reactor's thermal performance. Mechanical components must balance strength with minimal thermal resistance to optimize the reactor's heat transfer efficiency

* + 1. Functional design

The functional design of trickle-bed reactors involves defining key parameters such as single-channel length, the number of parallel channels, service-side specifications, and reactor head dimensions. The channel length is determined by calculating the packed bed length required to achieve the desired conversion, accounting for space for liquid redistributors in column reactors (Richardson et al., 2020), while multitubular reactors do not require redistributors due to smaller tube diameters. The active length is established by solving mass, energy, and momentum balances using a 1D plug flow reactor (PFR) model, incorporating bulk-to-catalyst transport, internal diffusion, and a modified Ergun equation for pressure drop (Ranade et al., 2011). The number of parallel channels is a crucial factor in multitubular setups, directly influencing productivity and heat dissipation. This parameter must be optimized to meet process specifications while maintaining effective heat management. The service-side hydraulic circuit, essential for jacketed columns and multitubular designs, is configured to manage heat transfer depending on whether the process is endothermic or exothermic. Standard tube-in-tube or shell-and-tube heat exchanger configurations are employed according to system requirements (Richardson et al., 2020). Lastly, the reactor head length is conservatively set equal to the reactor diameter to ensure uniform mechanical strength and operational stability across both column and multitubular setups.

* + 1. Verification step

The verification step, as illustrated in Figure 1, refines and finalizes the design by adjusting parameters to meet imposed constraints. In the input settings phase (Section 2.1), degrees of freedom include inlet conditions (e.g., temperature, pressure, and composition), reactor configuration (e.g., column, jacketed, or multitubular setups), geometric features (e.g., channel diameter), and process specifications (e.g., productivity, conversion, and yield). Transport properties related to flow regimes are also considered, while the catalyst type and kinetic model are assumed to be predetermined. Flow regime adjustments (Section 2.2) may involve selecting a new position on the flow map based on intrinsic physicochemical properties and previously established geometric parameters. Structural design aspects (Section 2.3) are typically fixed, as they depend on selected pressure loads, hydrostatic head, and material properties. However, adjustments to channel length may be necessary to account for hydrostatic pressure in vertical reactors. Material properties are treated as constants, selected based on trade-offs made during earlier stages. The functional design phase (Section 2.4) plays a crucial role in meeting process specifications, such as conversion and yield. If discrepancies arise between assumed parameters and specifications, iterative refinement is required. The design is validated once all degrees of freedom align with the initial assumptions and objectives. It is important to note that multiple configurations can be developed using this methodology. Additional multi-criteria frameworks, such as sustainability analyses, may also be used to guide decision-making for optimal solutions. However, such considerations are beyond the scope of this work.

* + 1. Economic analysis

To complete the overall evaluation of the proposed solution, the technical assessment has been complemented by an economic analysis (Andriani et al., 2024c). Specifically, both capital expenditures (CAPEX) and operational expenditures (OPEX) have been determined. For the CAPEX estimation, the purchase costs of both reactors and pumps were considered. The cost of acquiring basic carbon steel atmospheric equipment was determined and adjusted using material and pressure correction factors to account for specific process requirements. Regarding OPEX, the analysis accounted for electricity costs associated with the flow of both process and service fluids, as well as the cost of any service fluids, when applicable (Turton et al., 2018).

* + 1. Case study

To validate the proposed method, the catalytic liquid-phase hydrogenation of benzene to cyclohexane was selected as the case study. This reaction offers a practical approach for safe and efficient chemical hydrogen storage, as hydrogen can later be recovered via cyclohexane dehydrogenation to regenerate benzene, as depicted in Equation 1.

|  |  |
| --- | --- |
|  $C\_{6}H\_{6}+3H\_{2}\rightarrow C\_{6}H\_{12} ( ∆\tilde{H}\_{r}=-206.2 kJ/mol)$ | (1) |

A commercial nickel-alumina catalyst (Crosfield; Ni 16.6 wt%, surface area 108,000 m²/kg, pore volume 0.37 × 10⁻³ m³/kg, bulk density 860 kg/m³) was selected for the study (Toppinen et al., 1996). The catalyst's kinetic properties, as reported in the literature (Metaxas and Papayannakos, 2006), were utilized to model the reaction dynamics. The techno-economic analysis of benzene hydrogenation as a chemical hydrogen storage method plays a critical role in assessing the feasibility of centralized versus decentralized applications. Specifically, if the scale of a hypothetical plant required for the hydrogenation reaction demands a series of equipment rather than a compact, single-vessel design, a centralized approach is likely more practical. Conversely, evaluating the feasibility of a decentralized configuration for dehydrogenation requires a detailed assessment. In this case, a single large-scale facility could produce a hydrogenated liquid organic hydrogen carrier (LOHC), such as cyclohexane, off-site, while on-site dehydrogenation could be performed to meet specific hydrogen demand.

* 1. Results and discussions

Before presenting the results of the design architecture (Figure 1), the imposed boundary conditions are summarized in Table 1.

*Table 1. Common boundary conditions for all designed configurations.*

|  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Variable** | Tliq,in | Tgas,in | Pin | X | Y | xB,in | yH2,in | xH2,in | Hc | Ltot |
|  | [°C] | [°C] | [bar] | [kg m-2 s-1] | [kg m-2 s-1] | [-] | [-] | [-] | [m] | [kg s-1] |
| **Value** | 25 | 80 | 30 | 0.5 | 10 | 1 | 1 | Equilibrium | 6 | 70 |

Based on the values in Table 1, the liquid inlet temperature (Tliq,in) was selected to represent benzene storage at ambient temperature. The gas inlet temperature (Tgas,in) (Saebea et al., 2017) and inlet pressure (Pin) (Bornemann et al., 2024) were chosen based on the conditions of a hydrogen feed from a PEM electrolyzer, facilitating the storage of renewable electrical energy in the form of a liquid organic hydrogen carrier (LOHC). The reactor is assumed to operate under trickle-flow conditions, with pure liquid benzene introduced into the system. The liquid stream entering each reactor tube is assumed to have an equilibrium concentration of dissolved hydrogen, maintained by sufficient mixing in the reactor head. Table 2 summarizes the techno-economic results for the column (non-jacketed and jacketed) and multitubular configurations based on the boundary values specified in Table 1. For consistency, a pump efficiency of 0.75 has been assumed in all scenarios. Additionally, the process-side hydraulic channels are assumed to be constructed from stainless steel (i.e., AISI 316) due to hydrogen exposure, while the service-side channels and pumps are assumed to be made of carbon steel, as they are exposed only to water and benzene, respectively. In the economic assessment, no compressor has been included, as the operating pressure of the reactors has been assumed to match that of the electrolyzers.

*Table 2. Specific design conditions for all considered configurations.*

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
|  |  | **Column****(Non-jacketed)** | **Column****(Jacketed)** | **Multitubular****(Case A)** | **Multitubular****(Case B)** |
|  | **Configuration ID** | **S1** | **S2** | **S3** | **S4** |
|  | **Variable** |  |  |  |  |
| Geometricvalues | Total height[m] | 9.40 | 9.40 | 8 | 8 |
| Channel diameter [m] | 1.40 | 1.40 | 0.075 | 0.050 |
| Channel thickness [mm] | 17 | 17 | 1 | 1 |
| Number of channels [-] | 1 | 1 | 341 | 767 |
| Gas flow per channel [kg s-1] | 21 | 21 | 0.062 | 0.027 |
| Liquid flow per channel [kg s-1] | 70 | 70 | 0.205 | 0.091 |
| Serviceside | Service fluid | - | Water | Water | Water |
| Inlet service temperature [°C] | - | 25 | 25 | 25 |
| Inlet service pressure [bar] | - | 2 | 2 | 2 |
| Liquid service flow rate [kg s-1] | - | 90 | 200 | 200 |
| Shell diameter [-] | - | 1.47 | 2.00 | 1.97 |
| Shell thickness [mm] | - | 2 | 2 | 2 |
| Tube’s arrangement | - | - | Triangular | Triangular |
| Pitch [mm] | - | - | 96 | 65 |
| Number of baffles [-] | - | - | 10 | 10 |
| Outletvalues | Outlet temperature [°C] | 139.3 | 133.7 | 76.1 | 69.7 |
| Outlet pressure [bar] | 25.2 | 25.2 | 25.0 | 24.9 |
| Conversion [%] | 7.8 | 7.9 | 9.2 | 9.5 |
| Outlet service temperature [°C] | - | 27.5 | 38.7 | 41.2 |
| Economicindicators | CAPEX (k€ 2024) | 213 | 224 | 168 | 195 |
| OPEX (€/h 2024) | 46.8 | 58.7 | 73.3 | 73.3 |

Table 2 presents the analysis of five configurations: a single non-jacketed column (S1), a single jacketed column (S2), and two multitubular setups with varying tube diameters (S3 and S4). For the single-column configurations, temperature control of the reacting mixture proves inadequate, with a temperature variation of nearly 115°C. Efficient temperature regulation is crucial, particularly in exothermic processes, to prevent runaway reactions (Andriani et al., 2024d). Such large temperature fluctuations hinder hydrogen mass transfer from the gas to the liquid phase, limiting the reaction rate and reducing the maximum outlet conversion. When comparing the jacketed and non-jacketed columns, the cooling system remains ineffective due to the low ratio of external surface area to reaction volume, which prevents efficient heat dissipation.

In both the jacketed and multitubular configurations, water is selected as the cooling medium due to its non-fouling properties, high thermal conductivity, low cost, and inherent safety. The water mass flow rate is set to achieve a superficial velocity of approximately 1 m/s, balancing heat transfer efficiency with acceptable pressure drops on the shell side. The multitubular setup with a triangular tube arrangement enhances cooling performance, resulting in improved benzene-to-cyclohexane conversion. Decreasing the tube diameter further optimizes heat management; however, this increases the required number of tubes, necessitating a trade-off between equipment complexity and cost-effectiveness. Overall, with an active reactor length of 6 m under the specified conditions, a maximum conversion of approximately 10% is achieved. Reaching higher conversions with a single reactor is impractical. Even in the most thermally efficient configuration (S4), the temperature still rises by about 35°C, accompanied by a pressure drop of nearly 17%. Therefore, additional cooling and pumping systems, as well as multiple reactor stages, are necessary to maintain efficient fluid flow and mitigate severe mass transfer limitations. Effective thermal management remains a critical factor for exothermic reactions such as the hydrogenation of organic compounds, to prevent runaway scenarios.

From an economic standpoint, the capital expenditures (CAPEX) for equipment procurement, including reactor vessels and pumps, remain relatively consistent across the S1 and S2 configurations, while the S3 and S4 setups offers cost savings of approximately 25 % and 13 % considering the most expensive setup, respectively. However, significant differences arise in the operating expenditures (OPEX) among the various alternatives.

The presence of a service-side hydraulic circuit in certain setups leads to increased expenses for electricity and service water required for operation. This effect is particularly pronounced in the S3 and S4 configurations due to the high service fluid flow rates necessary for these designs. While the multitubular setups achieve an approximately 18% improvement in conversion efficiency compared to the single-column configurations, this gain is offset by a 30% to 35% increase in operating costs. Notably, this assessment considers a single piece of equipment rather than a series of reactive units. In a series configuration, operating costs could be amortized, as the primary energy demand is related to pressurizing reactants from storage to operating pressure. As a result, the conversion gains offered by the multitubular design may ultimately compensate for the higher OPEX. Nevertheless, a detailed evaluation of an equipment train configuration is necessary to validate these assumptions.

* 1. Conclusions and future directions

Hydrogen is increasingly recognized as a pivotal energy carrier for a sustainable future; however, its storage poses significant safety challenges due to the risk of forming explosive and flammable mixtures, as well as the extreme conditions required for high- and low-temperature storage. In this context, storing hydrogen in organic molecules, such as cyclohexane, offers a safer and more practical alternative, supporting the broader adoption of hydrogen-based technologies. This work presents a versatile and holistic design strategy for trickle-bed reactors aimed at hydrogen storage through the hydrogenation of benzene.

The proposed design approach is adaptable to both column (jacketed and non-jacketed) and multitubular configurations. Applied to the benzene hydrogenation case study, key insights emerge for large-scale implementation. Specifically, the column configurations demonstrate suboptimal heat management, with an average temperature increase of approximately 110°C, compared to 45°C for the multitubular setup. This substantial disparity in heat removal capability limits the overall conversion within a 6 m active reactor length, with a nearly 20% reduction in conversion efficiency from the most to the least effective configuration due to pronounced gas-liquid mass transfer limitations. Although the capital expenditures (CAPEX) for the four configurations are relatively comparable, the operating expenses (OPEX) tell a different story. The multitubular setups (S3 and S4) exhibit a 30% to 35% increase in OPEX compared to the single-column configurations (S1 and S2).

Achieving near-complete conversion requires the integration of intermediate cooling stages and pumping systems. Cooling the liquid mixture reduces mass transfer resistance, thereby promoting hydrogen absorption rather than stripping. Additionally, pumps and compressors are essential for maintaining efficient fluid flow and compensating for pressure drops across the reactor system. A detailed evaluation of the equipment train configuration is necessary to balance conversion efficiency gains against OPEX increases, ensuring the overall viability of the design. Given the substantial plant size required to achieve near-unity conversion rates, a centralized benzene hydrogenation station emerges as the most practical solution.

Future research will focus on developing a comprehensive process design to optimize benzene hydrogenation for industrial-scale applications. This strategy will include sustainability assessments that encompass techno-economic analysis, safety considerations, and environmental impact evaluations to identify the optimal balance among performance, cost, and safety. By addressing these factors holistically, the proposed approach aims to advance the safe and efficient integration of hydrogen storage solutions within the energy infrastructure. Eventually, further efforts will be devoted also the dehydrogenation section, aimed to recover the stored hydrogen from the involved organic materials.

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