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Optimisation of Synthetic Natural Gas Production via Direct Air Capture and Utilisation using Reduced Models under a Novel Trust-Region Funnel Method

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ABSTRACT

In this study, we propose a novel trust-region funnel (TRF) optimisation framework for process systems that integrate external black-box models, such as rigorous models, within equation-oriented (EO) formulations. The framework is applied to optimise a synthetic natural gas production process combining direct air capture and catalytic CO_2 conversion using dual-function material (DFM) technology, with the objective of minimising the total annualised cost. The problem is formulated in Pyomo and solved using IPOPT, treating the DFM reactor as an external black-box model. The TRF method achieves substantial improvements compared to published mixed-integer nonlinear programming and direct nonlinear programming approaches, reducing capture cost from 460 USD to 426 USD per tonne of CO_2. Key design improvements include reducing the number of DFM units per train by one-third and achieving a 22% reduction in DFM capital costs. These results highlight the TRF framework's ability to overcome numerical challenges and unlock economically superior designs for next-generation carbon capture and utilisation systems.

Keywords: Algorithms, Derivative Free Optimization, Catalysis, Carbon Capture, Reaction Engineering

INTRODUCTION

Modern challenges in process systems engineering (PSE) increasingly involve multi-scale optimisation problems aimed at rigorously assessing the performance and scalability of emerging technologies, such as carbon capture and utilisation (CCU) systems [1]. These problems are inherently hybrid, combining explicit equation-oriented (EO) glass-box process models with external, implicitly defined components, commonly referred to as black-box models. Black-box components typically originate from rigorous high-fidelity models (e.g., systems of computationally challenging nonlinear algebraic equations or ordinary and partial differential equations) or simulation packages, whose analytical structure is often unavailable or impractical to embed directly within EO formulations. When integrated into EO optimisation frameworks, rigorous high-fidelity models frequently introduce numerical instability and cause convergence issues in traditional nonlinear programming (NLP) solvers, while simulation packages cannot be incorporated as NLPs due to their model form [2].

Achieving convergence to first-order optimality in hybrid glass-box/black-box optimisation problems requires specialised solution strategies that explicitly account for the presence of external models. The classical trust-region filter method is a promising approach for solving such hybrid problems, but it faces significant challenges, including excessive numbers of expensive model evaluations and strong sensitivity to initialisation and algorithmic tuning parameters [3]. To address the limitations of NLP solvers and the trust-region filter method in handling rigorous high-fidelity models, this work proposes a trust-region funnel (TRF) optimisation framework for embedding rigorous external models within nonlinear process flowsheets. The TRF method extends the trust-region filter approach [4] by replacing the filter globalisation strategy with a funnel-based method [5]. This modification improves robustness, preserves convergence guarantees, and reduces reliance on

heuristic parameter tuning, enabling the optimisation of tightly coupled hybrid systems. A key feature of the proposed TRF strategy is its ability to efficiently utilise derivative information whenever available, thereby reducing the computational cost associated with external model evaluations. When analytic derivatives of black-box models are unavailable, the method remains applicable by employing derivative approximation at the cost of additional model evaluations. This flexibility makes the TRF method particularly well-suited for large-scale hybrid optimisation problems involving cyclic operation, implicit system behaviour, or computationally challenging and derivative-free sub-models.

Growing concerns regarding the long-term environmental risks and uncertainties associated with CO_2 storage have intensified interest in CO_2 utilisation pathways [5]. Among these, catalytic conversion of captured CO_2 into synthetic natural gas (SNG) using renewable hydrogen, commonly referred to as power-to-gas (PtG), has emerged as a promising option. This study demonstrates the application of the proposed TRF framework to optimise an integrated SNG production process based on direct air capture (DAC) and catalytic conversion using dual-function material (DFM) technology. The DFM reactor integrates CO_2 capture and methanation within a single unit [6]. In addition to the DFM unit, the process flowsheet includes upstream air intake and high-temperature solid oxide electrolyser (SOEL) units, and downstream heat-integration and separation systems.

The resulting problem is characterised by a large number of equality constraints and strong nonlinear coupling across multiple process units, making it computationally challenging for standard NLP solvers. To address these challenges, previous works [6][7] imposed restrictive assumptions, such as fixing key design and operating variables (e.g., adsorption time and DFM mass). While effective for numerical convergence, these restrictions significantly reduce the feasible design space, preventing identification of the true economic optimum.

In this work, we overcome these limitations by introducing additional degrees of freedom, allowing all process variables, including adsorption time and DFM catalyst weight, to vary during the optimisation process. The DFM reactor is treated as an external multi-output black-box component, embedded within an EO formulation, and the resulting hybrid optimisation problem is solved using the proposed TRF method. The results demonstrate that the TRF approach achieves robust and systematic optimisation of process systems that are otherwise difficult to optimise using traditional NLP techniques.

TRUST-REGION FUNNEL METHOD

The general form of the hybrid optimisation problem considered in this work can be expressed as:

$$\min_{w,z} \; f(w,y,z) \tag{1.1}$$

$$s.t.\; h(w,y,z) = 0 \tag{1.2}$$

$$g(w,y,z) \le 0 \tag{1.3}$$

$$y = t(w) \tag{1.4}$$

where f denotes the objective function, h and g represent equality and inequality constraints, and $t(w)$ is the rigorous (truth) model describing external black-box components. The functions f, g and h are assumed twice continuously differentiable. The variables w correspond to the inputs of the truth model, y are the associated external model outputs, and z denotes the remaining variables.

Trust-Region Subproblem

Trust-region methods iteratively solve a trust-region subproblem (TRSP) by replacing the truth model with a reduced model (RM). The RM is constructed to be sufficiently accurate within a neighbourhood of the current iterate, satisfying the κ-fully linear property as defined in [8]. The TRSP at iteration k is formulated as:

$$\min_{w,z} \; f(x) \tag{2.1}$$

$$s.t.\; h(x) = 0 \tag{2.2}$$

$$g(x) \le 0 \tag{2.3}$$

$$y = r_k(w) \tag{2.4}$$

$$\|u - u_k\| \le \Delta_k \tag{2.5}$$

where $x = [w,y,z]^T, r_k(w)$ denotes the RM at iteration k, u is a selected sub-vector from the variable vector $v = [w,z]^T$ that represents independent variables, u_k is the current base point, and Δ_k is the trust-region radius. The trust-region constraint restricts the step size in u to ensure that the RM provides a sufficiently accurate approximation of the truth model within the trust-region.

Reduced Model Construction

To enforce local consistency between the reduced and truth models, zero- and first-order correction terms are introduced whenever derivative information from the truth model is available or can be approximated. In this work, the RM is constructed as:

$$r_k(w) = \tilde{r}(w) + \big(t(w_k) - \tilde{r}(w_k)\big) + (\nabla t(w_k) - \nabla \tilde{r}(w_k))^T (w - w_k) \tag{3}$$

where $\tilde{r}(w)$ is a basis (or base surrogate model). In the present case study, $\tilde{r}(w) = 0$, such that the RM corresponds to a first-order Taylor approximation of the truth model about the current base point w_k. Although convergence guarantees hold for arbitrary choices of $\tilde{r}(w)$, algorithmic performance improves significantly as the accuracy of the $\tilde{r}(w)$ increases.

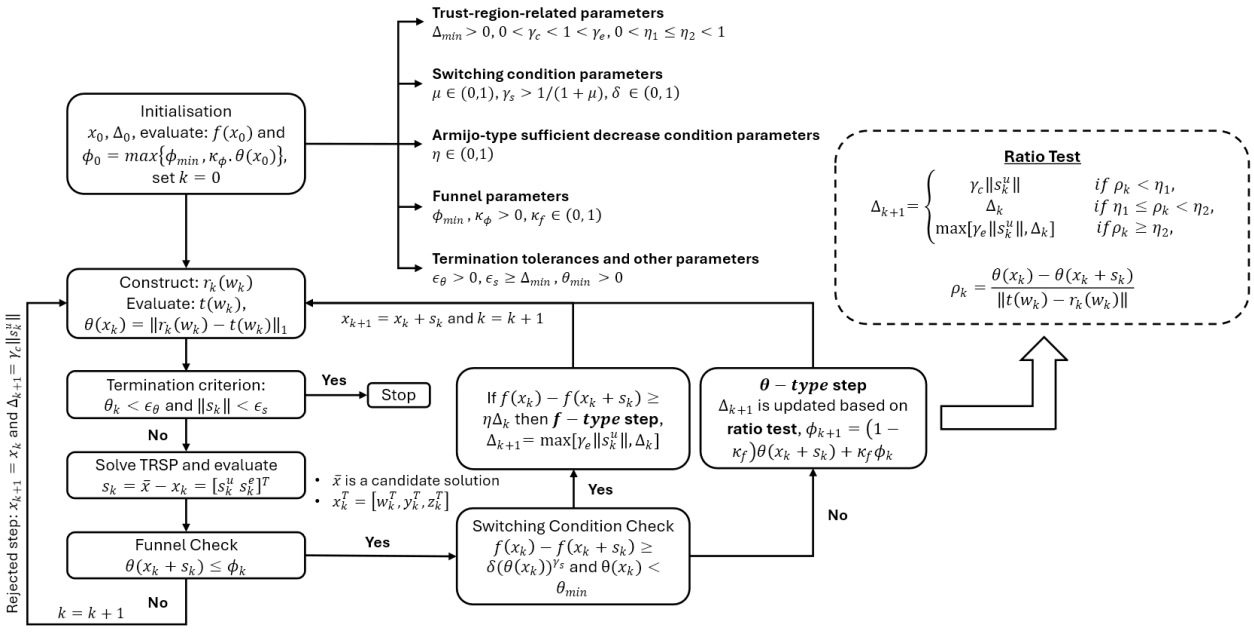

Figure 1: Trust-region funnel algorithm.

Funnel Globalisation Strategy

After solving the TRSP to obtain candidate solutions w_k^* and z_k^*, a funnel-based globalisation strategy is applied. The funnel enforces a progressively tightening upper bound ϕ_k on the RM approximation error θ, defined as:

$$\theta(w) = \|r(w) - t(w)\|_1. \tag{4}$$

Based on the agreement between predicted and actual reductions in the approximation error, quantified through a ratio test (see Figure 1 for details), and pre-defined funnel acceptance criteria, the trust-region radius is updated, and the candidate iterate is either accepted or rejected.

While the solution of each TRSP primarily targets improvement in the objective function, the funnel systematically drives successive iterates toward feasibility, enabling robust convergence to a locally optimal solution that satisfies first-order optimality conditions [9].

Algorithm Overview

The TRF algorithm proceeds iteratively as summarised in Figure 1. At initialisation, a base point x_0, initial trust-region radius Δ_0, and initial funnel bound $\phi_0 = \max\{\phi_{\min}, \kappa_\phi \cdot \theta(x_0)\}$ are set. At each iteration k, the reduced model $r_k(w_k)$ is constructed as per Equation (3) and the truth/rigorous model $t(w_k)$ is evaluated to compute the approximation error $\theta(x_k) = \| r_k(w_k) - t(w_k) \|_1$. If the termination criteria $\theta_k < \varepsilon_\theta$ and $\| s_k \| < \varepsilon_s$ are satisfied, the algorithm stops. Otherwise, the TRSP defined in Equations (2.1)-(2.5) is solved to compute a candidate step $s_k = [s_k^u, s_k^z]^T$, where superscripts u and z denote subvectors of the decision variables and the remaining variables, respectively.

The candidate iterate is then assessed through various sequential checks. First, a funnel check determines whether $\theta(x_k + s_k) \leq \phi_k$. If satisfied, a switching condition is evaluated: if sufficient objective reduction is achieved relative to the approximation error (i.e., switching condition) and trust-region size (i.e., Armijo-type sufficient decrease condition), an $f - type$ step is accepted and the trust-region radius is expanded. Otherwise, a $\theta - type$ step is taken, during which the funnel bound is updated as $\phi_{k+1} = (1 - \kappa_f)\theta(x_k + s_k) + \kappa_f \phi_k$, progressively tightening the feasibility envelope, and the trust-region radius is adjusted via the ratio test (as given in Figure 1). If the funnel check fails, the step is rejected and the trust-region radius is contracted.

This structure shares the switching condition and step-type logic of the classical trust-region filter method, but differs fundamentally in the globalisation strategy. The filter maintains an iteratively updated list of pairs (f, θ) and requires heuristic tuning of various problem-dependent tuning parameters. By contrast, the funnel can be interpreted as a filter with a single entry, i.e., the funnel width ϕ_k, updated only during $\theta - type$ iterations and initialised from $\theta(x_0)$ using a problem-independent parameter κ_ϕ. This eliminates the need to maintain non-dominated (f, θ) pairs or tune sensitive filter parameters, offering a key practical advantage. Importantly, this structural similarity enables convergence guarantees for the TRF method to be derived directly from trust-region filter theory [10].

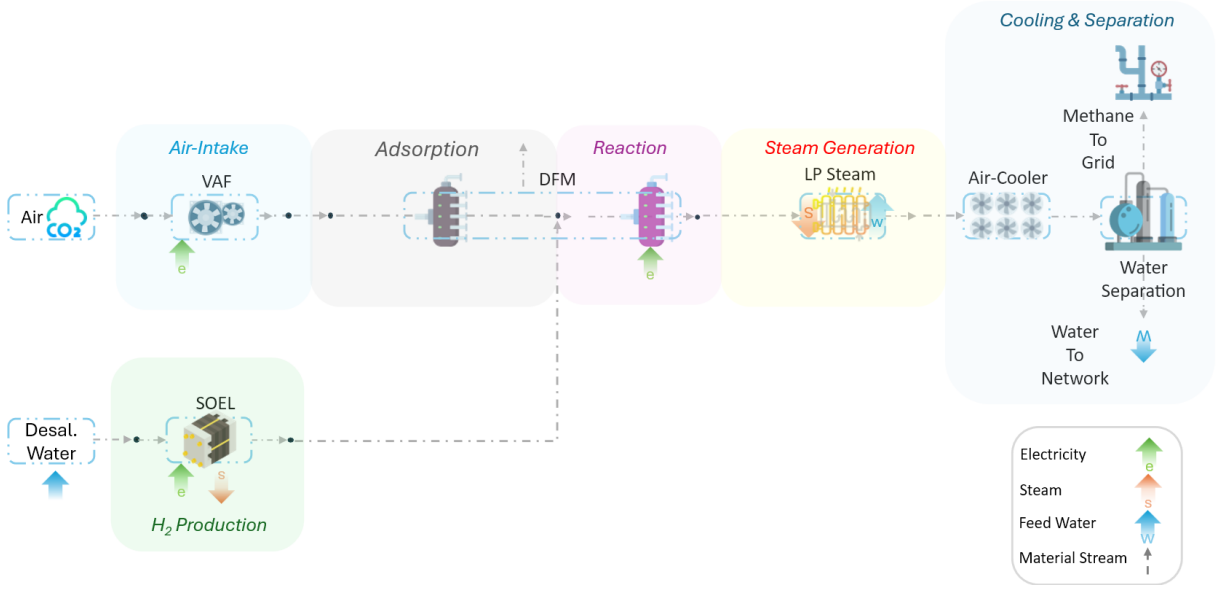

Figure 2: DFM DAC-to-SNG process flowsheet.

Convergence Guarantees

Under standard regularity assumptions, the proposed TRF method guarantees convergence to a first-order optimal point of the original hybrid optimisation problem, even in the presence of inexact or approximated derivative information from the external model [9][4].

PROCESS MODELLING

The optimisation objective is to minimise the total annualised cost (TAC) of the integrated process flowsheet shown in Figure 2. TAC accounts for capital and operating expenditures, including equipment sizing, material requirements such as water and DFM, and utility costs. The DAC process is assumed to be located within an industrial plant with access to electricity, cooling water, steam, and a boiler feed water network, ensuring the availability of required utilities. The mathematical models for the process units are adapted from [6][7].

Process Description

As illustrated in Figure 2, ambient air is introduced into the CO_2 capture unit via a vane axial fan (VAF) and directed into columns containing large tubes packed with honeycomb-shaped monoliths. These monolithic structures are coated with DFM, which facilitates CO_2 adsorption during the adsorption phase as the air stream passes through the monolith channels. The DFM formulation used in this study, $Ni^{(15\%)}Ru^{(1\%)}Na_2O^{(10\%)}/CeO_2$–$\gamma$ $Al_2O_3^{(74\%)}$, is based on published works [11][7]. During this cyclic process, CO_2 is captured on the DFM surface, followed by a short nitrogen purge to remove residual gases. Subsequently, a hot hydrogen stream at 350 °C, supplied by SOELs, is introduced to hydrogenate the adsorbed CO_2 in situ, producing methane. A final purge completes the cycle before repetition. All electrolysers operate at atmospheric pressure with 100% stoichiometric conversion of desalinated water to H_2 and O_2, simplifying the model by omitting downstream separation.

The hot gas generated during the cyclic operation is utilised in steam generators to produce low-pressure (LP) steam for export or internal heat integration. After heat recovery, the process stream is trim cooled using an air-cooler and directed to a flash drum for water removal. The resulting gas mixture primarily consists of methane, with residual amounts of water vapour, CO_2, and hydrogen. During high-conversion operation, unreacted hydrogen is minimal; however, when excess hydrogen is employed for temperature control or enhanced heat recovery, pressure swing adsorption (PSA) may be used for hydrogen recovery and recycle.

Reactor Modelling

The DFM reactor is modelled as a multi-output external black-box component within the nonlinear optimisation formulation of the flowsheet. Although the DFM model is equation-based, consisting of a coupled system of nonlinear algebraic equations that describe adsorption, hydrogenation, reactor geometry, energy balances, velocity, and pressure drop, its direct inclusion in the EO flowsheet results in non-convergence of classical NLP solvers, even with near-optimal feasible initialisation.

To ensure numerical tractability while preserving model fidelity, the DFM reactor is externalised as a black-box mapping, which returns product compositions and energy requirements as functions of the operating

conditions. The surrounding process flowsheet is formulated in Pyomo, and the TRSPs are solved using IPOPT.

RESULTS AND DISCUSSION

The case study considers a techno-economic optimisation of a DAC-DFM flowsheet designed to capture 10, 000 tonnes of CO_2 per year, consistent with recently reported industrial-scale benchmarks [6][7]. The proposed TRF framework successfully converged to a first-order stationary point in 38 iterations, as shown in Figure 3, which summarises the solver's performance.

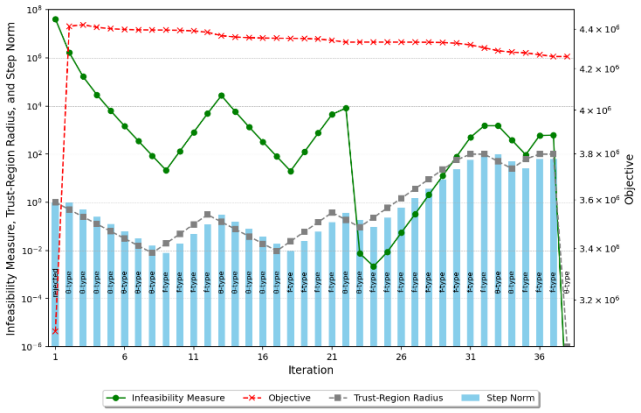

Figure 3. Performance metrics for the TRF solver.

Comparison of Optimisation Approaches

Key process metrics are summarised in Table 1, with an aggregated cost comparison presented in Figure 4. The study evaluates three optimisation approaches.

- The first approach uses a mixed-integer nonlinear programming (MINLP) formulation, where the DFM route, adsorbent mass, and adsorption time were fixed a priori [7].
- The second (direct NLP) optimisation approach converts the published MINLP flowsheet [7] to an NLP formulation while retaining fixed DFM weight and adsorption time to ensure solver convergence.

In the third (TRF-based grey-box) optimisation approach, the DFM reactor is treated as an external multi-output model, allowing both adsorbent weight and adsorption time to vary as decision variables.

As illustrated in Figure 4, the TRF framework achieves reduction in TAC compared to both the published MINLP and direct NLP solutions. While the published and direct NLP cases yield nearly identical results due to their fixed DFM design assumptions, the TRF solution reduces TAC by ~8%, demonstrating the economic benefits of relaxing restrictive design assumptions. The lower TAC is due to lower capital costs, while operational expenditures and revenues are observed to be similar for all optimisation approaches.

The observed improvement primarily stems from the optimisation of the adsorption–reaction unit design. Under the TRF framework, the number of DFM units per train is reduced from 16 to 6, while individual unit size and channel velocity are increased by ~2.9 and ~3.5 times, respectively. Adsorption time is also reduced from 1000 seconds to 626 seconds. These changes lead to a ~6% reduction in adsorbent weight and a corresponding ~23% decrease in DFM capital cost, without compromising capture targets or pressure-drop constraints. Note that the expensive catalyst sites in DFM reactor, particularly Ruthenium, result in significant capital expenditure in all optimisation approaches [7].

These results highlight a fundamentally different trade-off between reactor parallelisation (i.e., monolithic design), adsorption time, and adsorbent weight, systematically identified by the TRF algorithm. Unlike traditional approaches that rely on conservative over-design with multiple small parallel units, the TRF optimiser favours fewer, larger units operating at higher velocities and shorter cycle times. This design strategy reduces both equipment count and adsorbent inventory while maintaining overall process performance.

Economic and Practical Implications

This study illustrates that the algorithmic structure decisively influences not only whether a solution can be obtained but also its qualitative characteristics. By removing restrictive design and operational assumptions, imposed purely for numerical convenience, TRF method enables a faithful exploration of techno-economic search space and unlocks economically better designs, which is critical for emerging CCU and PtG systems to remain competitive. These systems inherently involve cyclic operation, strong nonlinear coupling, and implicit model formulations.

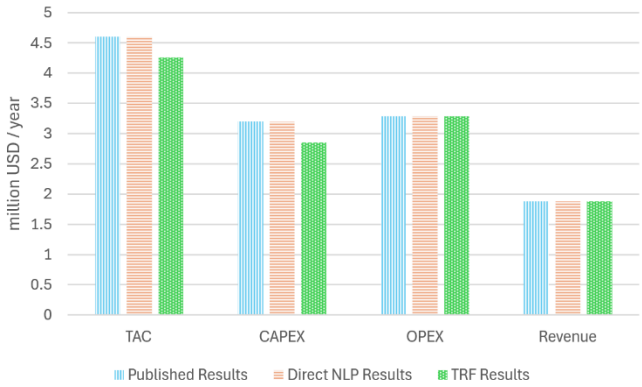

Figure 4. Economic comparison with published results.

Table 1: Optimisation results for the DFM-based process flowsheet (VAF: vane axial fan, SOEL: solid oxide electrolyser, DFM: dual-function material, LP: low pressure).

Variable Name	Unit	Published Results	Direct NLP Results	TRF Results
Air-Intake System: VAF				
Total capital cost	USD	705,100	705,030	705,030
Operational cost	USD/year	166,500	166,300	166,300
Number of fans	-	6	6	6
Fan power	kW	430	420	420
Hydrogen Production System: SOEL				
Total capital cost	USD	32,747,900	32,747,867	32,747,867
Operational cost	USD/year	3,115,800	3,115,738	3,115,738
Adsorption-Reaction Unit: DFM				
Total capital cost	USD	30,364,200	30,361,756	23,512,996
Operational cost	USD/year	0	0	0
Number of units in each train	-	16	16	6
Unit size (each)	m^3	22	22	63
Air velocity inside channels	m/s	2	2	7
Unit diameter	m	4	4	4
Unit height/length	m	2	2	5
Pressure drop in each unit	Pa	530	522	522
Total adsorbent weight	kg	28,350	28,343	26,678
Net CO_2 uptake	kmol/kg	0.000255	0.000255	0.000135
Steam Generation Level: LP				
Total capital cost	USD	54,600	54,555	54,555
Boiler heat transfer area	m^2	65	65	65
Steam production revenue	USD/year	26,000	25,950	25,950
Downstream Units				
Air-cooler total capital cost	USD	47,300	47,229	47,229
Air-cooler operational costs	USD/year	1,900	1,836	1,836
Separation vessel capital cost	USD	24,900	24,900	24,900
Separation vessel size	kg	340	339	339
Cost per tonne of CO^2 captured	USD/t_{CO^2}	460	460	426

CONCLUSION

This study demonstrates the effectiveness of the proposed TRF framework for optimising hybrid glass-box/black-box models in large-scale CCU processes, applied to a SNG production process integrating DAC and catalytic CO_2 conversion via DFM technology. The TRF method reduced TAC from 4.6 million USD to 4.26 million USD, achieving an ~8% decrease in the cost per tonne of captured CO_2. Key design improvements include reducing the number of DFM units per train (16 to 6), increasing unit size (22 m^3 to 63 m^3), and decreasing total adsorbent weight (28, 350 kg to 26, 678 kg), resulting in a 22% reduction in DFM capital costs. These results highlight the TRF framework's ability to overcome numerical challenges and deliver economically superior designs compared to traditional NLP/MINLP approaches. Future research will focus on embedding DFM reactor simulations based on partial differential equations within EO flowsheets to explore reactor dynamics and their interactions with the process. Moreover, integrating PSA as a downstream black-box unit can enable hydrogen recovery and recycling for enhanced process efficiency.

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