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Simultaneous Optimization of Design and Operating Conditions for RPB-based CO_2 Capture Process

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ABSTRACT

Although global efforts for CO_2 capture are underway, large-scale CO_2 capture projects still face economic risks and technical challenges. The Rotating Packed Bed (RPB) provides an alternative solution by mitigating location constraints and enabling a gradual increase in the scale of CO_2 capture through compact modular sizes. However, the main challenge in RPB-based CO_2 capture processes lies in the limited experience with implementing industrial-scale RPB processes. The intricate relationship between RPB unit design, operating conditions, and process performance further complicates the process-level analysis for scale-up. To address these challenges, we propose an optimization-based process design for RPB-based CO_2 capture. Leveraging rigorous process modeling and simulation, we aim to make simultaneous decisions on RPB unit design and operating conditions. Ultimately, our goal is to develop a cost-effective and optimal RPB-based CO_2 capture process, supported by comprehensive cost evaluations. This modularized and cost-effective approach is expected to facilitate rapid implementation and gradual scale-up, thereby reducing entry barriers to CO_2 capture technology for industries.

Keywords: Carbon Dioxide Capture, Process Intensification, Modelling and Simulations, Process Design, Technoeconomic Analysis

INTRODUCTION

In the ongoing efforts to combat climate change, CO_2 capture is expected to play a pivotal role for the foreseeable future. Among the available capture techniques, amine-based absorption stands out as a mature technology, with several industrial-scale CO_2 capture facilities in operation worldwide [1]. However, experiences such as the Petra Nova project, one of the largest industrial-scale CO_2 capture projects, have revealed the economic risks and technical complexities associated with building and operating such large-scale CO_2 capture processes. While economies of scale can benefit large-scale processes, the substantial initial cost and space requirement for vast CO_2 capture facilities make them less favorable investments [2]. Process intensification, particularly through technologies like the Rotating Packed Bed (RPB), offers a potential solution to address this hurdle.

RPB, as a form of process intensification, enhances mass transfer and unit throughput with the rotation of a packed bed. The increased centrifugal force widens the selection window for packing materials and facilitates the use of highly viscous solvents. This improved throughput can significantly reduce the required volume for column units, which typically account for approximately 50% of the capital expenditures (CAPEX), by up to 65% [3].

Driven by the potential benefits of RPB in shrinking mass transfer units, there have been numerous lab-scale experiments [4-6]. On the other hand, its application to industrial-scale CO_2 capture has been limited. Recent studies have explored RPB-based CO_2 capture on a scale of approximately 2200 tons per day (TPD)[7]. However, these studies have overlooked certain important factors, such as the pressure drop, in the RPB design, hindering the provision of realistic insights. Moreover, the overall lack of large-scale RPB-based process implementations poses challenges in extrapolating findings to larger and more practical scales. Notwithstanding the potential economies of scale, the practical viability of large-scale RPB units raises questions, especially considering that their characteristics are more aligned with small-to-medium scale applications.

The central question addressed in this research revolves around establishing a cost-effective design and optimal operating conditions for the RPB-based CO_2 capture process. The efficacy of applying RPB for CO_2 capture at an industrial scale remains uncertain, primarily due to a lack of experience in establishing such processes. Furthermore, limited insights exist regarding unit design guidelines and operating conditions for an industrial-scale RPB-based CO_2 capture process. While Agarwal et al. [8] presented a systematic RPB column design procedure, it is heuristics-based and can only address minimal design requirements to prevent flooding of the liquid or jet ejection of the vapor. Furthermore, the RPB column's design is contingent upon the initial assumptions about some process operating variables, e.g., the rotation speed of packing. The design of the RPB column influences mass transfer and hydraulic phenomena, impacting optimal conditions and overall performance. The inherent characteristics of the RPB demand iterative design and operating condition decisions, pointing to the limited effectiveness of the conventional sequential process design approach.

To address these limitations, we adopt a simultaneous optimization-based approach for both RPB design and operating conditions. Utilizing a comprehensive model for an RPB-based CO_2 capture process with a reference MEA solvent, we seek cost-effective RPB design and operating conditions for the compact CO_2 capture process. The envisioned modularized RPB-based CO_2 capture process aims to expedite implementation, enabling a phased scale-up of the CO_2 capture process and thus reducing entry barriers for the industry.

SYSTEM DESCRIPTION

RPB Column and Process Model

A process model for the RPB-based CO_2 capture process is constructed using the gPROMS custom modeling environment, building upon the foundation established in our prior publication [9]. The enhancement factor and effective surface area models have been updated for improved accruacty, incorporating a wider range of pilot plant operation data in this study. To ensure consistent insights into design and energy consumption, we seelcted the widely used MEA amine as the reference solvent. The increased centrifugal force in RPB units facilitates the application of high-viscosity solvents, prompting exploration into concentrated amine solvents, often enhanced with anti-degradation additives. MEA concentrations ranging from 30 to 75wt% MEA solvents are typical for RPB-based CO_2 capture, and we employed the eNRTL (electrolyte Non-Radom Two-Liquid) model with updated parameters for this broad range of MEA concentrations [10] as our thermodynamic model. The RPB column model adopts the two-film theory for the calculation of mass and heat transfer rates, complemented by an enhancement factor model.

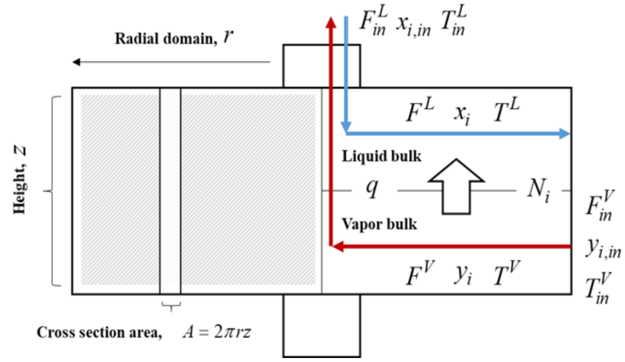

Figure 1. Conceptual scheme of RPB column model.

Figure 1 illustrates the conceptual scheme of the developed RPB column model, where vapor and liquid phases flow counter-currently along the radial axis, resulting in the formulation of the partial differential equation (PDE) with radial distribution:

$$\varepsilon^L \frac{\partial C_i^L}{\partial t} = -\frac{1}{2\pi rz}\frac{\partial (F^L x_i)}{\partial r} + a^I N_i \quad (1)$$

$$\varepsilon^V \frac{\partial C_i^V}{\partial t} = \frac{1}{2\pi rz}\frac{\partial (F^V y_i)}{\partial r} - a^I N_i \quad (2)$$

$$\varepsilon^L C_{tot}^L C_p^L \frac{\partial T^L}{\partial t} = -\frac{F^L C_p^L}{2\pi rz}\frac{\partial T^L}{\partial r} + a^I\big(h^I(T^V - T^L) + N_{H_2O}\Delta H_{H_2O}^{vap} + N_{CO_2}\Delta H_{CO_2}^{abs}\big) \quad (3)$$

$$\varepsilon^V C_{tot}^V C_p^V \frac{\partial T^V}{\partial t} = \frac{F^V C_p^V}{2\pi rz}\frac{\partial T^V}{\partial r} - a^I h^I (T^L - T^V) \quad (4)$$

Here, r_{inner}, r_{outer} and z represent the inner radius, outer radius, and height of the RPB unit, respectively. C and F denote concentration and molar flowrate, respectively. As the control volume changes along the radial axis, the continuity equations $F = 2\pi rzC_{tot}u$ hold for both vapor and liquid phases. The mass transfer rate of CO_2, N_{CO_2}, is calculated using the two-film theory with an enhancement factor as follows:

$$N_{CO_2} = K_{CO_2}^{overall}\left(P_{CO_2} - P_{CO_2}^*\right) \quad (5)$$

$$K_{CO_2}^{overall} = \frac{1}{\frac{RT^V}{k_{CO_2}^V} + \frac{He_i}{E_{CO_2} k_{CO_2}^L}} \quad (6)$$

Here, $K^{overall}$, P_{CO_2} and $P_{CO_2}^*$ represent the overall mass transfer coefficient, CO_2 partial pressure, and equilibrium CO_2 partial pressure, respectively. **Table 2** provides the transfer correlation and reaction models used for the column model. **Figure 2** illustrates the overall flowsheet of the RPB-based CO_2 capture process, encompassing the developed RPB model and additional process units within

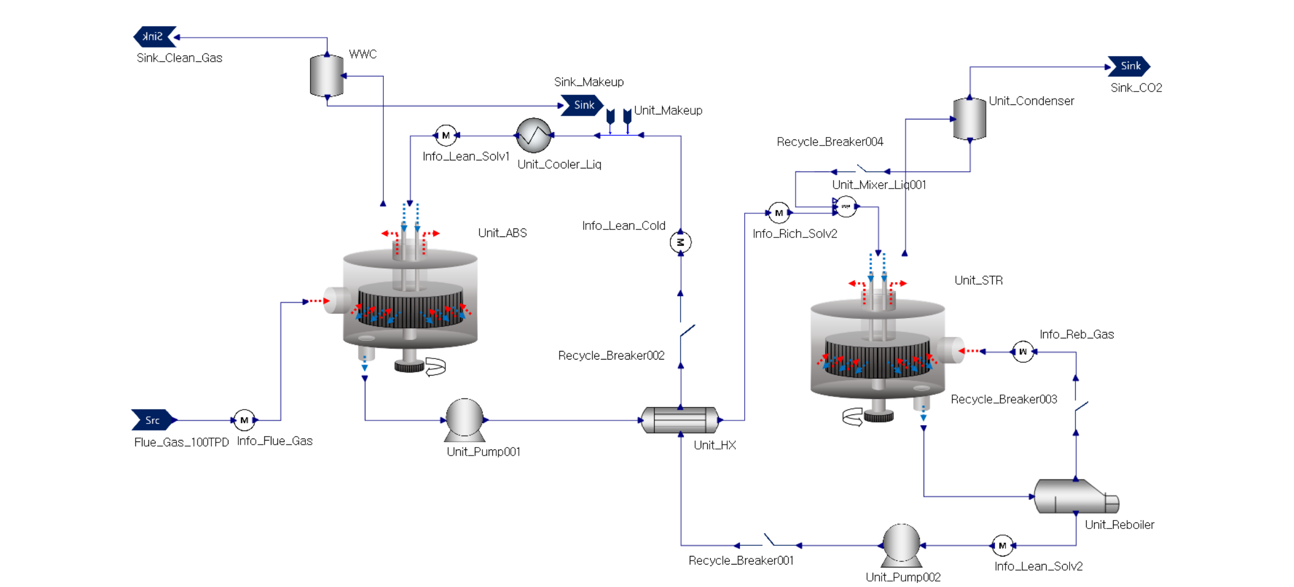

Figure 2: Overall flowsheet of RPB-based CO_2 capture process model (gPROMS).

the gPROMS simulation environment.

Table 2: Transfer correlation and reaction models

Variables	Model
Effective surface area (a^I)	Xie et al. [11]
Liquid transfer coefficient (k^L)	Tung et al. [12]
Vapor transfer coefficient (k^V)	Onda et al. [13]
Heat transfer coefficient (h^I)	Chilton-Colburn analogy
Enhancement factor (E_{CO_2})	Wellek et al. [14]
Reaction kinetics (k_{app})	Luo et al. [15]

Process Model Validation

Given the lack of comprehensive operational data for the overall RPB-based CO_2 capture process in existing literature, we subjected our developed process model to a validation process using operation data from separate absorber and striper pilot plants. We utilized pilot plant operation data from Jassim's (Run 1 to 16) [4] and Kolawole's study (Run 1 to 36) [16] for absorber column validation and Cheng's study (Run 1 to 12) for stripper column validation.

As shown in **Figure 3** and **Figure 4**, our validation results affirm the satisfactory fidelity of the developed RPB column model. The RPB model demonstrates robust predictive capabilities, with an error range of about ±10%p for CO_2 capture rate and ±20% for energy consumption. Even under a range of MEA concentration and various operating conditions, the RPB model maintains a mean absolute relative error (MARE) of 11.4 % and 9.2% for CO_2 capture rate and energy consumption, respectively.

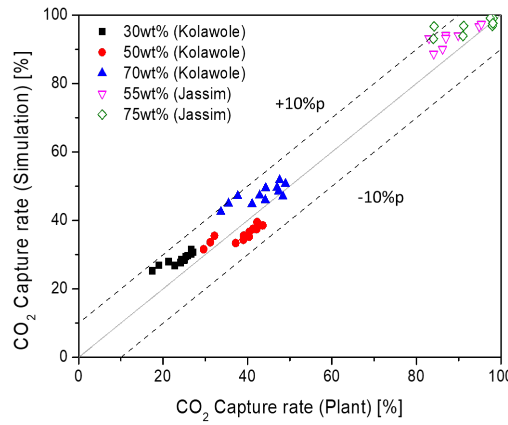

Figure 3. Comparison of pilot plant and process model (CO_2 capture rate).

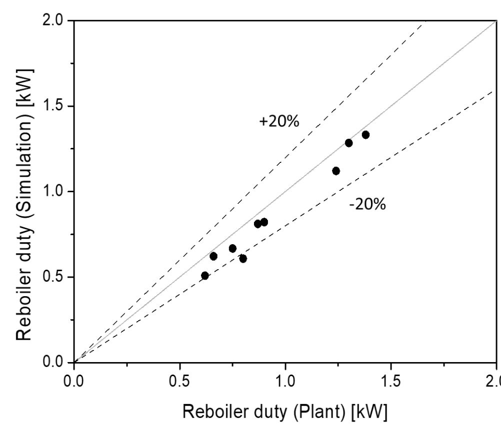

Figure 4. Comparison of pilot plant and process model (Energy consumption).

ANALYSIS OF LARGE-SCALE PROCESS

Target Scale and System

The implementation of an excessively large-scale RPB-based CO_2 capture process faces inherent challenges, given the quadratic increases in both rotational energy and pressure drop. The rotational nature of the RPB introduces escalating demands for momentum and centrifugal energy as the process unit size expands. In addition, the heightened angular acceleration and torque as the plant scale increase can require additional maintenance and safety concerns due to the intricate system control, increased mechanical strain on components, impact on the lifespan of process units, elevated risk of failures, and potential discomfort for operators [17, 18]. Consequently, the preference leans towards favoring an industrial-scale RPB-based process in the small-to-medium range, deviating from the conventional CO_2 capture process with fixed packed beds.

In this study, we explore a medium-scale process at 100 TPD CO_2 capture, specifically targeting flue gas from a coal-fired power plant. This choice aligns with a pragmatic compromise, as the flue gas flow rate for the 100 TPD scale equates to the flue gas from an approximately 6 MW scale power plant. **Table 3** shows the considered inlet flue gas stream condition in this study.

Table 3: Inlet flue gas stream condition

Variables	Value
Scale (TPD CO_2 capture)	100
Flow rate (kg/s)	5.94
Temperature (°C)	40
Pressure (bar)	1.1
Mole fraction, CO_2 (%)	14.5
Mole fraction, H_2O (%)	6.8
Mole fraction, N_2 (%)	76.6
Mole fraction, O_2 (%)	2.1

Simultaneous Optimization of RPB Design and Operating Condition

The highly coupled nature of RPB design and operating conditions often leads to local optima when a heuristic-based and sequential process design approach is applied. Consequently, the simultaneous determination of process unit design and operating conditions can offer a more cost-effective solution.

In our simultaneous design approach, an optimization problem is formulated to minimize the total annual cost (TAC) per the amount of captured CO_2, utilizing both RPB design parameters (d) and operating variables of the overall process (x) as decision variables. The formulation is as follows:

$$\min_{d,x} \frac{TAC}{\dot{m}_{CO_2,Captured}} = \frac{ACC+AOC}{\dot{m}_{CO_2,Captured}} \quad (7)$$

$$d \in [r_{inner,j}, r_{outer,j}, H_j] \quad (8)$$

$$x \in [F_{Solv}, T_{Reb}, P_{Str}, \omega_j] \quad (9)$$

$$\text{s.t.} \quad \eta_{Cap} \geq 90\% \quad (10)$$

$$T_{Reb} \leq 120℃ \quad (11)$$

$$0\% \leq \phi_{flood,j} \leq 80\% \quad (12)$$

$$r_{min,j} \leq r_{inner,j} \leq r_{outer,j} \quad (13)$$

$$H_j / r_{outer,j} \leq 1 \quad (14)$$

In the above formulation, ACC and AOC represent the annualized capital and operating cost, respectively. The index j represents the RPB units, encompassing absorber (Abs) and stripper (Str) columns. We employ the Lang factor method, based on the free-on-board (FOB) cost with a value of 5.93 for continuous process [19], to estimate the capital cost. Meanwhile the operating cost is calculated from the energy consumption using rigorous process simulation. The details on decision variables as summarized in **Table 4.**

Table 4: Considered decision variables

Process variables	Symbols
RPB inner radius (m)	$r_{inner,j}$
RPB outer radius (m)	$r_{outer,j}$
RPB height (m)	H_j
Solvent flowrate (kg/s)	F_{Solv}
Reboiler temp. (°C)	T_{Reb}
Stripping pressure (bar)	P_{Str}
RPB rotation speed (RPM)	ω_j

Constraints include a lower bound on 90% capture rate, an upper bound of 120°C on reboiler temperature to prevent thermal degradation of solvent, and a constraint on the 80% flooding condition to govern the RPB design parameters. Additionally, constraints on the minimum RPB inner radii [8] and mechanical recommendations for RPB design [20] are incorporated to ensure the reliability of RPB design.

Due to the absence of a specialized capital cost estimation model for industrial-scale RPB units in existing literature, we adopt a cost estimation model designed for centrifuges. The FOB purchase cost for the RPB unit is assumed to be the aggregate of costs associated with the rotation components (motor and rotor) and the packing bed. Leveraging cost models developed for centrifuges, the cost model unfolds as follows:

$$C_{RPB}^{FOB} = C_{Centrifuge}^{FOB} + V_{RPB}^{Packing} C_{RPB}^{Packing} \quad (15)$$

$$C_{Centrifuge}^{FOB} = \$6180 \cdot (D_{outer})^{0.94} \quad (16)$$

In the above equations, $V_{RPB}^{Packing}$ and $C_{RPB}^{Packing}$ represent the volume and cost of packing, respectively, with a packing price of $285/ft[3]. D_{outer}denotes the diameter of centrifuges (RPB in this study) in inches. For utility cost calculations, $8/GJ, $19.2/GJ, and $0.015/GJ are applied for steam, electricity, and cooling water, respectively.

RESULT AND DISCUSSION

The cost model developed for the simultaneous design approach enables a comparative analysis of CO_2 capture costs against the sequential heuristics-based design. In the heuristics-based method, RPB units were designed using Agarwal's RPB design procedure [8], and operating conditions were determined through an optimization problem aimed at minimizing energy consumption per captured CO_2 as follows:

$$\min_x E_{Cap} = E_{Steam} + C_{conversion}E_{Elec} \qquad (17)$$

The decision variables in eq (9) were employed, and a conversion factor of 1/0.4 was applied to align energy types.

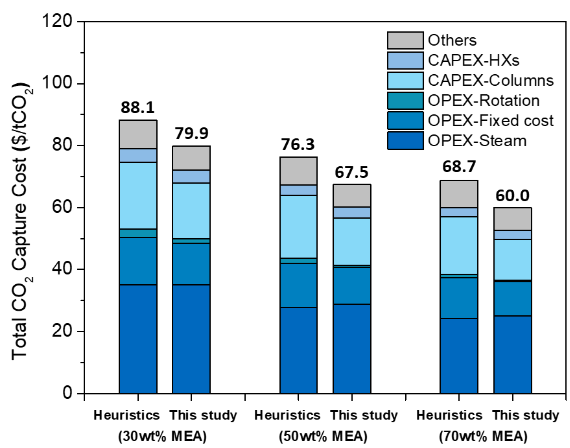

Figure 5. Comparison of CO_2 capture cost with heuristic and simultaneous design approach according to varying MEA concentrations.

Figure 5 illustrates the total CO_2 capture cost at varying concentrations of MEA solvent for both heuristics-based and simultaneous design approaches. The estimated total capture costs with RPB units range from $60.0 to 88.1/tCO$_2$, with the primary contributors being utility cost from steam, RPB column CAPEX, and fixed operation costs. Notably, the energy consumption by packing rotation does not constitute a significant portion of the overall cost. Given that the RPB CAPEX constitutes a large portion of the CO_2 capture cost, obtaining empirical data and developing appropriate cost estimation models will be a crucial for future scale-up studies of RPB-based processes.

The results reveal that employing the simultaneous decision results in an 9.4-12.7% reduction in CO_2 capture cost. This reduction is predominantly attributed to a decrease in column CAPEX, highlighting the effectiveness of incorporating RPB design parameters into the optimization problem as decision variables. The limitations of the sequential and heuristic design approach become more evident through the optimal design parameters and process variables shown in **Table 5** and **Table 6**. The optimized RPB design parameters and rotational speed exhibit significant differences in both approaches. The optimization results align with our understanding that maintaining the inner radius at a minimum is advantageous. However, the RPB design from the simultaneous solution shows the increased RPB height and reduced outer radius. Such a tall and stout RPB design is expected to contribute not only to reducing RPB CAPEX but also rotation energy and pressure drop, while maintaining the same unit throughput.

Table 5: RPB design and optimized process variables (Heuristics-based approach)

Variables	30wt%	50wt%	70wt%
ABS-Inner radius (m)	0.18	0.18	0.18
ABS-Outer radius (m)	1.31	1.28	1.19
ABS-Height (m)	0.36	0.30	0.28
STR-Inner radius (m)	0.10	0.10	0.09
STR-Outer radius (m)	0.35	0.29	0.23
STR-Height (m)	0.43	0.36	0.31
Solvent flowrate (kg/s)	21.2	12.5	9.1
Reboiler temp. (°C)	120*	120*	120*
Stripping pressure (bar)	1.86	1.58	1.08
ABS-RPM (rpm)	491	498	502
STR-RPM (rpm)	503	415	425

*Constraint active

Table 6: Optimized RPB design and process variables (Simultaneous optimization approach)

Variables	30wt%	50wt%	70wt%
ABS-Inner radius (m)	0.18*	0.18*	0.18*
ABS-Outer radius (m)	1.03	0.91	0.80
ABS-Height (m)	1.03	0.91	0.80
STR-Inner radius (m)	0.10*	0.10*	0.09*
STR-Outer radius (m)	0.27	0.21	0.18
STR-Height (m)	0.27	0.21	0.18
Solvent flowrate (kg/s)	21.7	13.8	10.0
Reboiler temp. (°C)	120*	120*	120*
Stripping pressure (bar)	1.88	1.62	1.13
ABS-RPM (rpm)	269	279	292
STR-RPM (rpm)	786	756	888

*Constraint active

Variations in MEA concentration also exert a significant influence on process performance and total capture costs. Increasing the MEA concentration from 30wt% to 70wt% results in notable savings in energy consumption and capture cost, ranging from 28.6 to 31.3% and 22.1 to 25.0%, respectively. This observed improvement is mainly attributed to the reduction in the amount of water evaporation heat in the reboiler. Furthermore, there is a notable reduction in the size of the RPB units, which can be attributed to the accelerated CO_2 absorption reaction rate associated with the increased amine concentration. While the widespread adoption of highly concentrated MEA solvent on an industrial scale is constrained by concerns about solvent degradation, the expanded solvent selection window offered by the RPB unit proves advantageous in saving energy consumption and reducing the total CO_2 capture cost.

The interdependence of rotational speed and RPB design is evident, as alterations in the RPB unit design necessitate adjustments in the rotation speed. Particularly in larger-scale processes, lowering the rotation speed becomes advantageous to counteract the quadratic rise in energy consumption with packing rotation.

CONCLUSION

The RPB-based CO_2 capture process is anticipated to find application across diverse industries owing to its enhanced processing capacity and reduced space requirements. To address the lack of process-level cost evaluation and a systematic design procedure for scaling up on this system, we undertook an exploration of a cost-effective capture process through modeling, simulation, and optimization. Initially, a first principle-based process model was developed and validated. By simultaneously optimizing RPB design and process operating conditions, we realized cost savings of 9.4-12.7% compared to sequential and heuristics-based design approaches. The estimated total CO_2 capture cost with RPB units ranged from $60.0 to 88.1 per ton of CO_2, varying with MEA concentration. The optimal RPB design found in this study, characterized by a taller height and shorter diameter, deviates from the heuristics-based approach. Given the strong correlation among RPB design, operating conditions, and process performance, our simultaneous design approach for RPB-based process holds promise for insightful process analysis across various scales in the future.

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