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Optimizing Sorbent Flow and Parameters in Calcium Looping Project integrated with CaO reactivation

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ABSTRACT

Achieving carbon neutrality and mitigating global warming demand effective management of anthropogenic CO_2 emissions. Calcium Looping (CaL) technology, characterized by its reversible carbonization and calcination reactions, presents a cost-effective and environmentally benign approach for CO_2 capture from coal-fired power plants. Integrating Concentrated Solar Heat (CSH) with CaL emerges as a promising avenue for cleaner production. Addressing calcium sintering issues in CaL projects, this study employed a wet precipitation method for Precipitated Calcium Carbonate (PCC) production to reactivate deactivated calcium material. Balancing the adsorbent flow rate and reactor temperature is paramount for cost-effective carbon dioxide capture. Consequently, this study conducted an optimization analysis of adsorbent flow rate and reactor parameters to enhance carbon capture efficiency and economic viability. Kinetics of the carbonator and calciner were modeled in Aspen. The findings revealed optimal temperatures of 600°C for the carbonator and 900°C for the calciner, considering system cost and CO_2 capture efficiency. The study determined that the molar flux ratios of recycled absorber to CO_2 in flue gas and supplemental absorber to CO_2 in flue gas yielding the lowest cost are 0.09 and 4, respectively. The findings furnish valuable insights for configuring flow rates and temperatures in project construction, thereby fostering the commercialization of carbon capture initiatives, and advancing carbon neutrality goals.

Keywords: Calcium Looping, CSH, CaO Reactivation, sorbent flow rate, parameter optimization

NONMENCLATURE

Abbreviations	
CaL	Calcium Looping
CCS	Carbon Capture and Storage
CSH	Concentrate Solar Heat
PCC	Precipitated Calcium Carbonate

1. INTRODUCTION

In response to the growing imperative to achieve carbon neutrality and mitigate global warming, it is essential to manage and reduce anthropogenic CO_2 emissions [1]. The power sector is currently advancing carbon capture and storage (CCS) technologies and renewable energy sources to achieve this goal [2]. However, due to the protracted energy transition, coal will remain a dominant energy source for the foreseeable future [3, 4]. Consequently, CCS technologies are crucial in reducing CO_2 emissions from coal-fired power plants.

Among various CCS solutions, calcium looping (CaL) technology has garnered significant attention [5]. This method involves a reversible reaction of carbonation and calcination of calcium and carbon dioxide at approximately 600°C and 900°C, respectively, to capture CO_2 [6]. Compared to other CCS methods, CaL is cost-effective and non-toxic due to its use of natural limestone as a precursor [4, 7]. Additionally, the carbonation reaction releases high-quality energy, which, when combined with a heat recovery system, can effectively minimize energy waste [7, 8]. These advantages position CaL technology as a promising retrofit for coal-fired power plants to reduce CO_2 emissions.

Extensive research has been conducted on the feasibility of using CaL technology to capture CO_2 from combustion flue gas [6, 7, 9, 10]. For instance, Alonse et al. [11] demonstrated the effectiveness of a fluidized bed carbonator reactor in capturing CO_2, while Arias et al. [12] successfully applied CaL technology in a 1.7 MW demonstration power plant. Despite its promise, CaL technology faces challenges such as the large amount of heat required for calcining $CaCO_3$, often provided by coal combustion, which generates pollutants like sulfur and

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ash [12, 13]. These pollutants can reduce capture efficiency and negatively impact the environment, making the technology unsustainable.

A current research focus is the use of concentrated solar heat (CSH) to supply the necessary heat for calcination. This approach combines the benefits of clean energy and reduces harmful byproducts, leveraging the thermal energy storage capability of CaL to address solar energy intermittency. Studies by Ortiz et al. [7] and Bayon et al. [14] have affirmed the feasibility of integrating CSH with CaL through detailed analyses of reactors, process parameters, and sorbent materials.

However, several issues persist with CaL technology, notably calcium sintering at high temperatures, necessitating the replenishment of deactivated $CaCO_3$ [15]. This requirement poses a sustainability challenge due to the extensive limestone extraction needed. Furthermore, impurities in limestone can adversely affect CO_2 capture efficiency. Research into alternative materials is therefore crucial. Inspired by Owais et al.'s [16] work on precipitated calcium carbonate (PCC) production from slag in the iron and steel industry, treating sintered CaL materials with ammonium chloride solution and wet precipitation offers a promising reactivation method, reducing dependence on fresh limestone.

Many studies have extensively explored the technical and economic feasibility of the Calcium Looping (CaL) process, demonstrating its potential for high efficiency and cost-effectiveness [4, 11]. However, current research has primarily focused on the design and analysis of individual reactors within CaL technology, lacking comprehensive investigations into the effects of sorbent flow rates and reactor parameters. The circulation and replenishment flow rates of the sorbent, along with specific reactor parameters, profoundly impact the CO_2 capture performance and the overall energy demand of the process. Poor design decisions in these areas can result in lower efficiency and higher costs. Furthermore, with the introduction of sorbent reactivation processes and CSH technology in this study, variations in sorbent flow rates and the sizing of the CSH system will also affect the economic viability of the system. Thus, a thorough study of these parameters is essential to determine the precise requirements for integrating CaL with power plants.

Therefore, the objective of this study is to assess the impact of integrating flow parameters and reactor parameters in CaL technology on the capture efficiency and economic performance of the project. To achieve this objective, this study proposes to develop an optimization analysis of sorbent flow rate and reactor temperature to evaluate the effects of parameter variations on the technical and economic performance of CaL projects. This analysis will provide a foundation for selecting optimal flow rates and parameters for the construction of such projects, thereby promoting the commercialization of carbon capture projects and contributing to carbon neutrality. Such advancements are critical for the power sector to meet stringent emissions targets and play a pivotal role in the global effort to combat climate change.

2. MODEL DESCRIPTION

The scheme comprises three primary components: the CSH system, the CaL system, and the sorbent reactivation system. Figure 1 depicts the connections among these components. This study does not delve into the complexities of power plant design; instead, it uses flue gas from a 660 MW supercritical coal-fired power

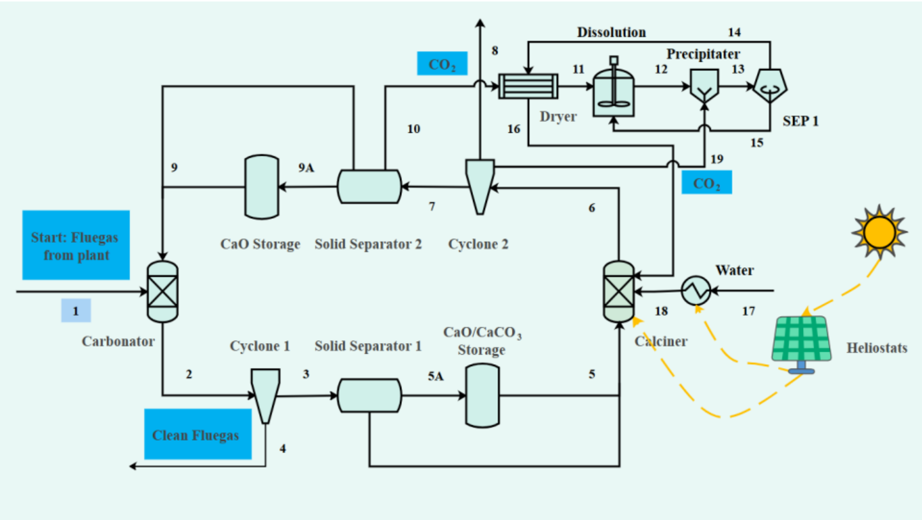

Figure 1 The connections for the combined model

plant in India as input for the CaL project [17]. The main volumetric components of the flue gas were 14.9% CO_2, 7.7% H_2O, 3.6% O_2, and 73.8% N_2 [17]. Sulphur dioxide (SO_2) was excluded from this analysis due to its negligible concentration. Furthermore, the study does not address the compression and storage of captured CO_2 or the application of heat released during the CaL process.

The mass and energy balance proposed system were calculated using Aspen Plus 12 with the following key assumptions: (i) simulations were based on the Peng-Robinson thermodynamic model; (ii) the system was operated under ideal steady-state conditions, excluding factors such as heat dissipation and pressure drop; and (iii) optimization of specific design and process details, including the reactor and solids storage, was beyond the study's scope.

The main elements of the CaL system include the carbonator, calciner, cyclone separator, and storage tank. The carbonator captures and fixes CO_2 from the flue gas, operating typically at 550-650 °C. The calciner facilitates the thermal decomposition of calcium carbonate ($CaCO_3$) at 800-950 °C, releasing high concentrations of CO_2 and regenerating the sorbent. The carbonator and calciner were modeled using the RSTOIC model in Aspen Plus. Gas-solid separation was achieved using a cyclone separator, assumed to operate at 100% efficiency with negligible solids loss.

Ideally, the CaL process would operate without additional sorbent materials. However, high-temperature calcination deactivates the sorbent, necessitating periodic renewal to maintain CO_2 capture efficiency. The maximum CO_2 capture efficiency is determined by the molar flow rate ratio of recirculating sorbent to CO_2 in the flue gas and the molar flow rate ratio of supplemental sorbent to CO_2 in the flue gas [10].

$$\eta_{\text{cap,max}} = \frac{F_0 f(1-b)}{F_{\text{CO}_2}(F_0/F_{\text{R}}+1-\text{f})} + \frac{F_{\text{R}}}{F_{\text{CO}_2}} \cdot b \qquad \text{Eq.1}$$

Where, F_0 denotes the molar flow rate of the supplemental sorbent, and f and b are characteristic parameters of the sorbent (0.77 and 0.17, respectively).

According to Eq.1, increasing the two parameters enhances CO_2 capture efficiency but also increases the system's size and energy demand. Excessive supplemental calcium flow can destabilize the system, while high circulating solids can inactivate most of the remaining calcium, reducing carbonation efficiency. Optimal recirculation and supplemental absorber flow rates result from trade-offs involving reactor size, energy input, and cost-related parameters, analyzed in this study with a focus on cost optimization. Detailed optimization is presented in Section 3.

The maximum average conversion of CaO is not constant. Reaction temperature significantly impacts CaO conversion in the CaL system, with CO_2 capture efficiency varying with carbonization temperature due to reaction rate changes and reverse reactions. Excessive calcination temperatures can deactivate the absorber and increase energy demand. Factors such as temperature and residence time of the carbonation and calcination reactions affect CaO conversion. Kinetic modeling of the carbonator and calciner was conducted to determine optimal operating parameters using the RCSTR model in Aspen. Table 1 lists the operating parameters and values analyzed in the kinetic model.

Table 1 The operating values in the kinetic model

Reactor	Parameter	Range	Step
Carbonator	Temperature (℃)	500-700	10
	CO_2 fraction (%)	5-20	5
	Residence time (min)	0-15	3
Calciner	Temperature (℃)	850-950	25
	Sorbent cycle times	1,2,3,5,20	-

The kinetic parameters of the carbonation reaction are described by a reaction model based on the grain model, emphasizing that sorbent conversion is mainly controlled by the fast chemical reaction phase, with the diffusion phase exerting less influence on conversion. The specific reaction rate of carbonation at atmospheric pressure is expressed in Eq.2 [18, 19].

$$\text{R}(s^{-1}) = 56k_{carb}\left(P_{\text{CO}_2} - P_{\text{CO}_2,\text{eq}}\right)^n S \qquad \text{Eq.2}$$

Where, 56 is the molar mass of CaO, g/mol; n is the reaction order, which is 1st order in this study; P_{CO_2} is the partial pressure of CO_2, atm; $P_{\text{CO}_2,\text{eq}}$ is the partial pressure of CO_2 at equilibrium, atm; S is the surface area of CaO, m^2/g; and k_{carb} is the rate constant of the carbonation reaction, which is analyzed using the Arrhenius Equation for computational analysis [19].

$$k_{carb} = k_0 \exp\left(-\frac{E}{R_g T}\right) \qquad \text{Eq.3}$$

Where, k_0 is the pre-exponential factor, taken as 30 kmol/(atm·m²·s) in this study; E is the activation energy of the reaction, 20.3 kJ/mol; R_g is the ideal gas molecular constant, 8.314 J/(mol·K); and T is the temperature of the reaction (K).

The reaction temperature of CO_2 at equilibrium can be calculated by using the Eq.4 proposed by Baker [20].

$$P_{\text{CO}_2,\text{eq}} = 10^{7.079-8308/T} \qquad \text{Eq.4}$$

As for the kinetics of the calcination process, the modified grain model proposed in the literature was used to describe it in this study [21].

$$\frac{\text{d}f_{\text{calc}}}{\text{d}t} = k_{calc}(1-f_{\text{calc}})^{0.67}\left(\text{C}_{\text{CO}_2,\text{eq}} - \text{C}_{\text{CO}_2}\right) \qquad \text{Eq.5}$$

Where, f_{calc} is the fraction of total $CaCO_3$ decomposed in the calcination reaction; k_{calc} is the rate constant of the calcination reaction, with a chosen pre-exponential factor of $2 \cdot 10^6$ $m^3/(kmol \cdot s)$ and an activation energy of 112.4 kJ/mol; the last two symbols represent the concentration and equilibrium concentration of CO_2 in the gas phase, $kmol/m^3$, calculated using Eq.6 [22].

$$\mathrm{C}_{\mathrm{CO_2,eq}} = \frac{P_{\mathrm{CO_2,eq}}}{RT} \qquad \text{Eq.6}$$

The CSH system mainly consists of a fixed-sun mirror field providing heat for the calciner reaction. This field was not modeled in Aspen; instead, dimensions were calculated based on calciner heat and heat transfer efficiency. Under light conditions, the CSH supplies heat to the calciner. Excess energy from the heliostat field is stored as calcium oxide in a storage tank at the calciner exit and released to the carbonator under non-light conditions. Storage tank dimensions were derived from theoretical calculations by Bayon et al. [14] The energy transfer efficiency between the heliostat field and calciner can be calculated below, according to Zhang et al. [23].

$$\eta_{solar} = \eta_{ref} \cdot \eta_{clc} \cdot \eta_{fld} \cdot \eta_{int} \cdot \eta_{rec} \qquad \text{Eq.7}$$

Where, η_{ref} represents the reflectivity of the heliostat field to sunlight; η_{clc} is the degree of heliostat cleanliness; η_{fld} is the efficiency of the heliostat field; η_{int} is the aperture interception of sunlight and η_{rec} represents the efficiency of the calciner in absorbing energy from the heliostat field. Table 2 summarizes the parameters used in the simulation of the CSH system.

Table 2 CSH system parameters [23,24]

Parameter	η_{ref}	η_{clc}	η_{fld}	η_{int}	η_{rec}	DNI
Value	93%	94%	78%	98%	85%	500W/m2

Waste solids are regenerated through a process similar to PCC production, involving purge dissolution, CO_2 bubbling carbonization, filtration, washing, and drying. Unlike PCC production from slag, the waste solids here are mainly pure CaO, making 100% dissolution and wet carbonization efficiency a reasonable assumption. Specific reaction characteristics of the regenerated sorbent require further testing, which is beyond this paper's scope.

3. ECONOMIC MODEL

As shown in Eq.1, the maximum CO_2 capture efficiency is determined by the molar flow rate ratio of the circulating sorbent to the CO_2 in the flue gas and the molar flow rate ratio of the reactivated sorbent to the CO_2 in the flue gas. Thus, these two sorbent molar flow rates play a critical role in determining the overall system size, energy requirements, and, consequently, project costs. In this investigation, the optimization objective revolves around the cost for achieving CO_2 capture throughout the entire system lifecycle. The optimal sorbent circulating sorbent flow rate F_{R} and reactivated sorbent flow rate F_0 were determined to minimize the cost of capture while upholding a CO_2 capture efficiency of 90%.

The capital cost of the carbonator, calciner, and CSH are the most significant parts of the project cost [4]. In addition, the material cost of the sorbent in the project is also important since we use the sorbent reactivation process to replace the purchase of fresh limestone. Additionally, the costs of the storage tank, cyclone, and piping are included, but these can be disregarded as they are negligible compared to the reactor costs. Therefore, for the simplicity of the study, only the carbonator cost, calciner cost, CSH cost, and sorbent cost are considered in the cost of this part.

The costing calculation method outlined in the literature employs a reasoned approach for assessing the calciner/carbonator reactor. It treats the reactor analogously to a boiler in a circulating fluidized bed, determining construction costs based on heat input/output. Regarding sorbent costs, the model considers two primary components, encompassing the initial sorbent purchase and transport cost of limestone and the production cost of reactivated sorbent. The limestone price is assumed as 25 €/t. The average limestone transportation distance and price are assumed as 500 km and 0.1 €/(t·km). The equipment cost equations for this calculation are summarized in Table 3. Importantly, the costs derived from this model are not indicative of the actual CO_2 capture cost but are utilized solely for determining the optimal sorbent make-up flow rate.

Table 3 The equipment cost equations

Equipment	Cost Function (M€)
Carbonator [4]	$599.57 \cdot (\frac{Q_{out}[MW]}{1527})^{0.67}$
Calciner [4]	$628.47 \cdot (\frac{Q_{in}[MW]}{2514})^{0.67}$
CSH [25]	$120 \cdot 10^{-6} \cdot A_{helio}[m^2]$
Sorbent reactivation [26]	$15.58 \cdot \left(\frac{\dot{\mathrm{m}}[Mtons/year]}{6}\right)^{0.7}$

4. RESULTS AND DISCUSSION

Figure 2 displays the refined parameters for the carbonator and calciner. Figures 3(a) and 3(b) illustrate the relationship between CO_2 capture efficiency and carbonator temperature, considering different CO_2 concentrations and residence times. The efficiency of CO_2 collection shows an initial increase and subsequently

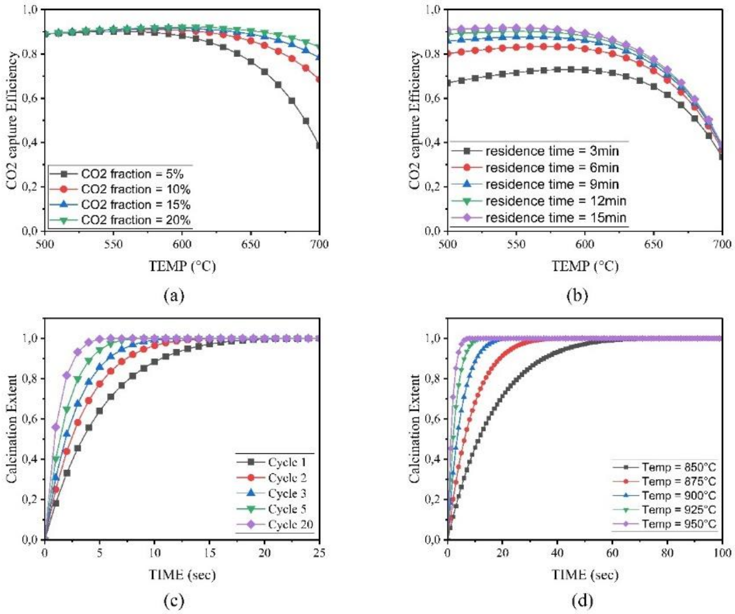

Figure 2 The refined parameters for the carbonator and calciner

decrease as the temperature rises, especially beyond 650 °C. This is due to the accelerated reaction rates and subsequent reverse reactions. Figure 2(a) demonstrates that the turning point corresponds to a gradual rise in temperature and an elevated level of CO_2 concentration. Meanwhile, Figure 2(b) demonstrates enhanced capture efficiency when the residence period is extended, but the benefits become less significant after 9 minutes.

Figure 2(c) demonstrates that when the number of adsorbent cycles increases in the industrial cycle, the calcination time reduces. This phenomenon occurs as a result of sintering at elevated temperatures, which causes the formation of aggregated adsorbent surfaces that exhibit reduced efficiency in capturing CO_2. Figure 2(d) depicts the degree of calcination of the new adsorbent at various temperatures, demonstrating that higher temperatures result in shorter calcination durations. Excessively high calcination temperatures can deactivate the absorber and markedly increase energy demand. Thus, to attain optimal capture efficiency, it is advisable to maintain temperature ranges of 550-600 °C for carbonization and 850-900 °C for calcination. Additionally, a residence time of 9 minutes in the carbonator is deemed suitable.

Figure 3 presents the relationship between CO_2 capture efficiency and the molar flow rate ratio of the circulating absorbent to the CO_2 in the flue gas and the reactivated absorbent to the CO_2 in the flue gas. An increase in either of the two ratios leaded to a rise in the active absorbent flow rate into the carbonator, partially counteracting the reduction in activity caused by sintering and thereby enhancing the CO_2 capture efficiency. However, excessively high make-up flow rates might disrupt reactor mass balance, complicating the system’s operation. Moreover, Martinez et al. indicated that carbonator and calciner costs were correlated with their heat duty, and excessive supplemental flow rates can elevate calcination heat requirements, further escalating costs. Thus, from practical and economic standpoints, achieving 100% carbon capture efficiency is

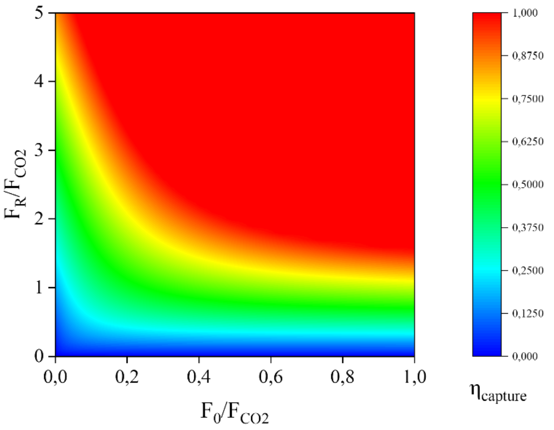

Figure 3 Relationship between CO2 capture efficiency and the molar flow rate ratio of the circulating absorbent to the CO2 in the flue gas and the reactivated absorbent to the CO2 in the flue gas

unfeasible. Consequently, in this study, seven combinations of flow parameters yielding a 90% carbon capture efficiency were selected from Figure 3, and their corresponding carbon capture costs were calculated and presented in Table 4.

The results were obtained with carbonator and calciner temperatures set at 600°C and 900°C, respectively. The data presented in Table 4 revealed that as the number of circulating absorbents increases, the requirement for absorbents to be calcined decreases. Consequently, both the cost of the calciner and the cost of materials decreased, resulting in an overall cost reduction despite the increased cost in the carbonator due to the cooling of more high-temperature absorbents to 600°C. Specifically, the cost of the calciner decreased from Combination 1 to Combination 2 because there was a reduction of 1/3 in the amount of $CaCO_3$ to be reactivated, leading to a decrease in the heat requirement and the associated cost of the calciner. Beyond Combination 5, increased circulating absorbent raises heat demand due to temperature differences between the calciner and carbonator. The diminished heat from the $CaCO_3$ calcination reaction failed to offset this disparity, leading to an increased size of the calciner and CSH costs. Weighing these factors, Combination 5 emerged as the most cost-effective within the system, considering total costs, chosen as our model input.

Table 4 Cost for CO_2 capture according to different absorbent flow rates

Parameters			Cost, M€				
Combination	F_R/F_{CO2}	F_0/F_{CO2}	Calc	Carb	CSH	Sorbent	Total
1	2.0	0.36	549	24	1085	81	1739
2	2.5	0.24	534	26	1042	59	1661
3	3.0	0.18	523	27	1029	46	1625
4	3.5	0.13	529	28	1028	38	1623
5	**4.0**	**0.09**	**531**	**30**	**1032**	**29**	**1622**
6	4.5	0.05	533	31	1038	23	1625
7	5.0	0.02	536	33	1047	16	1632

The correlation between reactor temperature and cost was also explored. Using carbonization and calcination temperatures of 600°C and 900°C respectively as basic situation, Figure 4 shows the relationship between system cost and calcination/carbonization temperatures. The results indicated that costs could be reduced by appropriately lowering the calcination temperature and increasing the carbonization temperature. The selection of this temperature range was predicated on a 14.9% CO_2 concentration in a 660 MW plant; hence, similar decisions should be based on the specific characteristics of the flue gas when considering reaction temperatures. Our study offers a new perspective on design considerations for commercial carbon reduction projects.

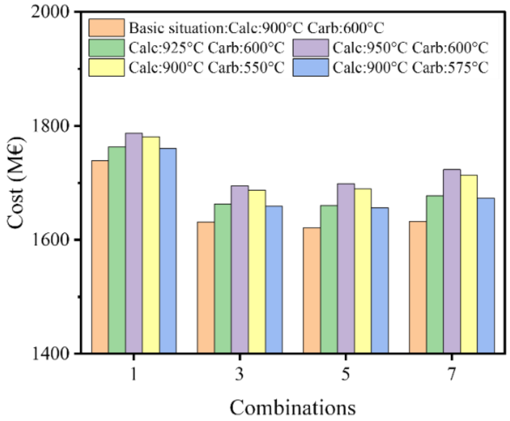

Figure 4 The relationship between cost and reactor temperatures against different absorbent flow rate settings

5. CONCLUSIONS

This manuscript addresses the challenge of high-temperature calcium sintering in Calcium Looping (CaL) technology by integrating calcium reactivation with CaL and employing Concentrated Solar Heat (CSH) for cleaner production and circular economy. Through Aspen kinetics simulations of the carbonator and calciner and economic modeling of the project, valuable insights into optimizing adsorbent flow rates and reactor parameters were gained.

The findings underscore the intricate balance necessary for optimal CaL system performance. Key parameters influencing system performance, such as carbonator and calciner temperatures, were identified, offering practical guidance for system design and operation. Optimum capture efficiency was achieved by maintaining carbonization temperatures between 550-600°C and calcination temperatures between 850-900°C, with a recommended residence time of 9 minutes in the carbonator.

Furthermore, the analysis revealed trade-offs among adsorbent circulation rate, reactor temperature, and system cost. By exploring various flow parameter combinations, the study demonstrated how optimization strategies can markedly reduce costs while preserving high capture efficiency. Specifically, the lowest-cost molar flow ratios of recycled sorbent to CO2 in flue gas and supplemental sorbent to CO2 in flue gas were found to be 0.09 and 4, respectively, for flue gas from a 660 MW coal-fired power plant.

Moreover, investigating the correlation between reactor temperature and system cost revealed opportunities for cost optimization through temperature adjustments, with carbonation and calcination temperatures of 600°C and 900°C minimizing costs for the targeted power plant.

In conclusion, this study enhances our understanding of CaL technology and its potential in achieving carbon neutrality. By addressing optimization challenges, CaL emerges as a cost-effective and sustainable solution for CO2 capture in coal-fired power plants, thus playing a pivotal role in advancing carbon neutrality objectives.

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