## A SYNERGY MODEL OF MATERIAL AND ENERGY FLOW ANALYSIS FOR THE CALCINATION PROCESS OF GREEN PETROLEUM COKE IN ROTARY KILN

#### by

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The main objective of this paper is to establish a mathematical framework to analyze the complex material and energy performance of the calcinations process based on the fundamental mass and energy conservations. The synergy degree of vital order parameters was defined and evaluated to assess the status and order of the calcination process. Furthermore, the synergy model for resource utilization and energy saving has also been developed. The results show that the energy efficiencies of the drying kiln, rotary kiln, incinerator, and the cooler are 63.574%, 37.709%, 76.782%, and 74.758%, respectively. Meanwhile, the synergy degree of the whole calcination system is determined as 0.507. Based on the result, several suggestions were proposed to improve the resource utilization, energy-saving and synergy performance. Assessing the performance of suggested improvements, the synergy degree was re-evaluated and recorded a substantial enhancement up to 0.809. The present work provides valuable insights and comprehensive analysis tool for assessing the performance and potential optimization of the calcination process.

Key words: petroleum coke, rotary kiln, calcination process, synergy degree, resource utilization, energy saving

## Introduction

In the aluminum electrolytic industry, calcined petroleum coke (CPC) and coal tar pitch are the raw material to make prebaked carbon anode for the aluminum cell application to produce aluminum [1]. Meanwhile, as the final solid residue of crude oil distillation refineries [2], green petroleum coke (GPC) could be used as the unique material to manufacture CPC through calcination process. Furthermore, aiming to improve the density, mechanical strength, conductivity and chemical stability of raw material, GPC needs to be heated up to remove moisture, volatiles and ash [3]. Although the traditional vertical shaft kilns are still commonly employed in China [4], the rotary kiln has become widely adopted for the calcination process, due to its high level of automation level and superior productivity and operating environment. Nevertheless, with the ever rising energy cost, large-scale materials and energy consumption have hindered the development of the calcination industry [5]. To improve the efficiency of

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this significant procedure, it is crucial to evaluate and optimize the synergy of the material and energy flow during the production process.

Recently, several research works have been carried out studying the calcination process. Firstly, some researchers focused on the final CPC quality. Bayram et al. [6] combined the discrete element method with the advanced 3-D imaging technique to investigate the influence of packing behavior of calcined cokes. Their study concluded that the low average sphericity has a direct impact on increasing the friction in the system. Heintz [7] performed an experimental investigation on the effect of calcination rate on the overall petroleum coke properties. It is found that a slower calcination rate is essential to produce regular or even premium quality CPC. Secondly, a number of studies focused on the characterization of the flow field, mass and heat transfer during the calcination process. Mastorakos et al. [8] adopted the Monte-Carlo method coupled with the finite-volume method to model the radiative heat transfer and energy conservation in the rotary flow. On the other hand, Xiao et al. [9] establish a numerical framework based on the CFD technique to characterize the distribution of residual moisture and volatiles in the petroleum coke, and the flow features of the gas and solid phases of calcination process. In addition, Elkanzi [10] also established a model to evaluate the material and energy balance of the rotary kiln for GPC calcination and verified the result against actual industry operation data.

From the perspective of thermodynamics, the calcination process of GPC in the rotary kiln is considered as an open, non-equilibrium, irreversible, non-linear and complex system. Therefore, the detailed theory of material and energy flow analysis and synergy is crucial to improve the resource utilization and energy saving in manufacturing process [11, 12]. Usually, researchers applied three sub-models to analysis the energy utilization of the system, including material and energy flows model [13, 14], synergy model [15] and exergy analysis model [16]. Filkoski *et al.* [17] analyzed five different options of waste heat utilization on exhaust gases from a shaft kiln. Similarly, Dolianitis *et al.* [18] applied a 3-D computational model to study the energy efficiency of glass furnace. In the past, research works on material, and energy flow or synergy model has been focused mainly on the metallurgical process [19, 20], construction industry [21] and cement industry [22]. Limited studies have been directed towards the calcination process. Therefore, it is urgently needed to understand the material and energy flow in calcination process to optimize the production yet minimize the energy consumption.

This paper presents a comprehensive methodology to analyze the material and energy flow in the calcination process based on our previous research works. Despite the work by Yu *et al.* [23] focusing on the smelting process of the steel belt sintering-submerged arc furnace, the subject matter of the present work has a distinct feature of process flow, method of resource utilization and energy saving, physical reaction, chemical reaction, *etc.* Consider the complex process, an advanced mathematical model for the mass and energy flow in the calcination process has been developed. Additional model modifications were also implemented considering the mass and energy flow stored in the control unit and generated within the control unit. New order parameters, together with the new formulation and evaluation principle, were also proposed to reflect the distinguishing features of the calcination process. Furthermore, analysis of resource utilization and energy saving in the process has been performed based on the synergetic theory. A synergy degree (SD) has been proposed to evaluate the coordination degree. Possibly improvements of the calcination process are also discussed.

#### Methodology

Like other metallurgical processes, material flow is mainstream of the system while the energy flow is the driving forces of the calcination process. Therefore, it is necessary to set up the mass and energy models to analyze the performance of material and energy flow of the calcination process in the rotary kiln. Nevertheless, traditional energy analysis methods cannot comprehensively reveal the interaction between material and energy flow or clearly describe the details of energy transformation within the system [24]. For better understanding the calcination process, the concept of synergy between material and energy flow is firstly introduced. The procedure structure considered in this article is expressed as follows. Firstly, the material balance of the four equipment is evaluated based on the desired of cooled CPC production capacity. Secondly, the energy balance can then be calculated by means of adjusting the independent variables of each equipment. Furthermore, a subsystem,  $IG_m$ , are extracted and in which various order parameters are defined, so as to study the synergy of  $IG_1$  and  $IG_2$ . At last, the SD of the calcination process can be calculated, which is an important parameter for evaluating the order and cooperation degree of the system.

## Description of the calcination process

The calcination process of GPC in the rotary kiln has been studied in the past years. A schematic of the process is shown in fig. 1. The process consists of four major equipment, including the drying kiln, the rotary kiln, the incinerator, and the cooler. After removing the moisture by the drying kiln, dried GPC is then undergone the calcination process in the rotary kiln. Afterwards, it is then cooled to a temperature below 80 °C by the cooler (CCPC). Meanwhile, the flue gas from rotary kiln, consisting of un-burnt particulates and volatile matter, is further processed in the incinerator for complete combustion. The high heat content in the flue gas is recovered in the waste heat recovery boiler (WHRB) for energy saving.



Figure 1. A schematic of the calcination process

## Synergy model of the process

Considering the complexity of the system, one should notice that each of the subsystems is closely coupled instead of mechanical superposition in some other simple systems. Although each subsystem has its own functions and characteristics, the subsystems conflict with each other sometimes, in order to promote the whole calcination process. Thus, the synergetic theory is applied to analyzes the calcination process for fine-tuning the system state from disorder to order. It is noteworthy that the material flow,  $Sub_1$ , level and energy flow,  $Sub_2$ , level are introduced to study the system. The fluctuations of material and the critical energy point affecting the system state are defined as the order parameters.

The order parameters exhibit two opposite characteristics in relation to the material and energy flow of the calcination system [25]. Consequently, the efficacy coefficient is defined to quantify the influence of order parameters based on the order to the subsystem, which is given by:

$$EC_{m}(V_{m,n}) = \begin{cases} \frac{V_{m,n} - V_{m,n}^{ll}}{V_{m,n}^{ul} - V_{m,n}^{ll}} & \text{for } V_{m,n} \text{ with positive effect} \\ \frac{V_{m,n}^{ul} - V_{m,n}}{V_{m,n}^{ul} - V_{m,n}^{ll}} & \text{for } V_{m,n} \text{ with negative effect} \end{cases}$$
(1)

where  $EC_m(V_{m,n})$ ,  $V_{m,n}$ , m, and n denote the efficacy coefficient, the order parameters, the indices of the subsystem, and the indices of the order parameters in the corresponding subsystems, respectively. Furthermore,  $V_{m,n}^{ul}$  and  $V_{m,n}^{ll}$  are the upper and lower limit of the order parameters when the system is operating at steady-state.

Meanwhile, the stability of the system depends not only on the individual values of the order parameters, but also on the combined effects of all parameters. Hence, the order degree, *i.e.*  $OD_m(IG_m)$ , is defined to describe the subsystem state, which is expressed:

$$OD_m(IG_m) = \sum_{n=1}^{N} [\omega_{m,n} EC_m(V_{m,n})]$$
<sup>(2)</sup>

where  $OD_m(IG_m)$  and  $\omega_{m,n}$  mean the order degree of the subsystem and the standard weight of efficacy coefficient, respectively. When  $OD_m(IG_m) = 1$ , it represents that the subsystem is at the perfectly order state. In contrast, if  $OD_m(IG_m) = 0$ , the subsystem is operating at the most disorderly state. In addition the entropy method [26] is used to calculate the standard weight, which is:

$$\omega_{m,n} = \frac{1 - S_{m,n}^{\text{en}}}{N - \sum_{n=1}^{N} S_{m,n}^{\text{en}}}$$
(3)

where  $S_{m,n}^{en}$  is the entropy value of the order parameter, which can be evaluated from the following formulation:

$$S_{m,n}^{en} = -\frac{1}{\ln K} \sum_{k=1}^{K} (\theta_{m,n}^k \ln \theta_{m,n}^k)$$
(4)

where *K* denotes the sample number of order parameter and  $\theta_{m,n}^k$  is the standardization value which is given by:

$$\theta_{m,n}^{k} = \begin{cases} \frac{V_{m,n}^{k} - \min_{K} \left\{ V_{m,n}^{k} \right\}}{\max_{K} \left\{ V_{m,n}^{k} \right\} - \min_{K} \left\{ V_{m,n}^{k} \right\}} & \text{for } V_{m,n} \text{ with positive effect} \\ \frac{\max_{K} \left\{ V_{m,n}^{k} \right\} - V_{m,n}^{k}}{\max_{K} \left\{ V_{m,n}^{k} \right\} - \min_{K} \left\{ V_{m,n}^{k} \right\}} & \text{for } V_{m,n} \text{ with negative effect} \end{cases}$$
(5)

In order to analyzes the synergy of the whole calcination process, it is necessary to integrate the order degree of each subsystem. Therefore, the geometric averaging method is used to evaluate the SD of the overall system, which can be expressed:

$$SD = M \sqrt{\prod_{m=1}^{M} OD_m(IG_m)}$$
(6)

## Results

# Material and energy flows analysis of the major equipment

Based on the operational data and boundary conditions, together with mathematical model of mass and energy flow, the results of mass and energy balance calculation of the major equipment are summarized in tabs. 1 and 2.

Ν	lass balance	of drying kiln		Mass balance of rotary kiln				
$M_{ m DK,IN}$ [th <sup>-1</sup> ]		$M_{\rm DK,OUT}$ [th <sup>-1</sup> ]		$M_{ m RK,IN}$	$[th^{-1}]$	$M_{ m RK,OUT}$ [th <sup>-1</sup> ]		
$M_{\rm DK,GPC}$	11.427	$M_{ m DK,FG}$	8.176	$M_{ m RK,NG}$	0.052	$M_{\rm RK,ICGPC}$	0.529	
$M_{ m DK,HA}$	6.973	$M_{\rm DK,DGPC}$	10.224	MRK,DGPC	10.582	$M_{ m RK,FG}$	14.598	
				$M_{ m RK,CA}$	12.499	$M_{\rm RK,CPC}$	8.006	
Total	18.400	Total	18.400	Total	23.133	Total	23.133	
Ν	Aass balance	of incinerator		Mass balance of cooler				
$M_{\rm IC,IN}$ [th <sup>-1</sup> ]		$M_{\rm IC,OUT}$ [th <sup>-1</sup> ]		$M_{ m CL,IN}$ [th <sup>-1</sup> ]		$M_{ m CL,OUT}$ [th $^{-1}$ ]		
$M_{\rm RK,ICGPC}$	0.529	MIC,RGPC	0.358	M <sub>CL,SW</sub>	4.003	M <sub>CL,DT</sub>	0.006	
$M_{ m RK,FG}$	14.598	MIC,FG	44.203	M <sub>CL,LA</sub>	1.283	$M_{\rm CL,FG}$	5.286	
<i>M</i> IC,CA	29.512	$M_{\rm IC,DT}$	0.078	MRK,CPC	8.006	$M_{\rm CL,CCPC}$	8.000	
				Mcl,ccwi	98.700	Mcl,ccwo	98.700	
Total	44.639	Total	44.639	Total	111.992	Total	111.992	

Table 1. Mass balance of major equipment

## Synergy model evaluation of the calcination process

In order to ensure a stable calcination process, it is necessary to establish a synergy model and several important order parameters based on the operation of material flow, Sub<sub>1</sub>, and energy flow, Sub<sub>2</sub>. The order parameters are adopted to analyze resource utilization and energy saving during the calcination process. In the economic perspective, it is desired to have a

high product yield of GPC while minimizes the carbon loss to reduce the resource consumption. The resource efficiency could be further enhanced by maintain a high recovery rate of gas and solid waste. It is also noteworthy that the continuity degree of the process also poses a profound contribution to the overall time efficiency and labor cost. In terms of promoting the overall energy saving, it is vital to minimize the external energy consumption of the CPC. Obviously, scavenging energy from the waste gas and waste solid could also reduce the overall energy consumption. The order parameters,  $V_{m,n}$ , for the material flow, Sub<sub>1</sub>, are defined as follows.

The product yield of GPC  $V_{1,1}$  is defined as the ratio of the mass production rate of the cooled CPC (CCPC) to the total mass input of the GPC which is given by:

$$V_{1,1} = \frac{M_{\text{CL,CCPC}}}{M_{\text{DK,GPC}}} 100 \tag{7}$$

E	nergy balance	e of drying kil	n	Energy balance of rotary kiln				
$E_{\rm DK,IN}$ [GJh <sup>-1</sup> ]		Edk,out [GJh <sup>-1</sup> ]		Erk,in [	GJh <sup>-1</sup> ]	Erk,out [GJh <sup>-1</sup> ]		
$E_{\rm DK,GPC}$	0.197	$E_{\rm DK,FG}$	1.026	Erk,ngch	2.615	Erk, ICGPC	0.651	
$E_{\rm DK,HA}$	4.667	Edk,dgpc	0.635	$E_{\rm RK,NG}$	0.003	$E_{\rm RK,FG}$	13.624	
		Edk,meh	3.092	Erk,dgpc	0.183	$E_{\rm RK,CPC}$	15.522	
		Edk,dhl	0.111	Erk,ca	0.315	$E_{\rm RK,MEH}$	1.346	
				$E_{\rm RK,VMCH}$	26.888	$E_{\rm RK,DHL}$	9.532	
				$E_{\rm RK,SCH}$	0.642			
				$E_{\rm RK,CCH}$	10.029			
Total	4.864	Total	4.864	Total	40.675	Total	40.675	
Energy balance of incinerator			Energy balance of cooler					
$E_{\rm IC,IN}$ [GJh <sup>-1</sup> ]		EIC,OUT [GJh <sup>-1</sup> ]		Ecl,in [GJh <sup>-1</sup> ]		Ecl,out [GJh <sup>-1</sup> ]		
Erk, ICGPC	0.651	$E_{IC,RGPC}$	0.067	$E_{\rm RK,CPC}$	15.522	Ecl,ccpc	0.497	
$E_{\rm RK,FG}$	13.624	$E_{\rm IC,FG}$	64.377	Ecl,sw	0.419	Ecl,swvh	10.867	
$E_{\rm IC,CA}$	0.744	$E_{\rm IC,DT}$	0.149	$E_{\rm CL,LA}$	0.026	$E_{\rm CL,DT}$	0.002	
$E_{\rm IC,VMCH}$	47.377	$E_{\rm IC,DHL}$	0.659	Ecl,ccwi	10.329	$E_{\rm CL,FG}$	0.236	
Eic,sch	0.026					Ecl,ccwo	14.461	
Eic,cch	2.830					Ecl, dhl	0.233	
Total	65.252	Total	65.252	Total	26.296	Total	26.296	

Table 2. Energy balance of major equipment

Similarly, the carbon loss  $V_{1,2}$  is defined as the combustion rate of the fixed carbon mass in the rotary kiln to the total mass of the fixed carbon in the dried GPC, which is expressed:

$$V_{1,2} = \frac{M_{\rm RK,FCC}}{M_{\rm DGPC,FC}} 100 \tag{8}$$

where  $M_{\text{RK, FCC}}$  and  $M_{\text{DGPC, FC}}$  denote the combustion rate of the fixed carbon mass in the rotary kiln and the total mass of the fixed carbon in the dried GPC.

The recovery rate of waste gas  $V_{1,3}$  and solid  $V_{1,4}$  are the crucial order parameters that have a significant effect on the resource utilization which are:

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$$V_{1,3} = \frac{M_{\rm RK,FG} + M_{\rm IC,FG}}{M_{\rm DK,FG} + M_{\rm RK,FG} + M_{\rm IC,FG} + M_{\rm CL,FG}} 100$$
(9)

$$V_{1,4} = \frac{M_{\rm IC,RGPC} + M_{\rm IC,DT} + M_{\rm CL,DT}}{M_{\rm RK,DGPC}} 100$$
(10)

Continuity degree of process  $V_{1,5}$  is defined as the ratio of the total production hours to the total working hours of the calcination process which is given by:

$$V_{1,5} = \frac{\Sigma t_{\rm P}}{\Sigma t_{\rm W}} 100 \tag{11}$$

where  $\Sigma t_P$  and  $\Sigma t_W$  denote as the total production hours and the total working hours of the process, respectively. The total working hours,  $\Sigma t_W$ , is:

$$\Sigma t_{\rm W} = \Sigma t_{\rm P} + \Sigma t_{\rm L} + \Sigma t_{\rm M} + \Sigma t_{\rm B} \tag{12}$$

where  $\Sigma t_L$ ,  $\Sigma t_M$ , and  $\Sigma t_B$  are the total latency hours, maintenance hours and breakdown hours of the process, respectively.

On the other hand, the order parameters,  $V_{m,n}$ , for the energy flow, Sub<sub>2</sub>, are introduced further in the text. The external energy consumption of CCPC  $V_{2,1}$  is defined as the required external thermal energy consumption per unit ton of CCPC needed, which is given by:

$$V_{2,1} = \frac{E_{\rm RK,NGCH} + E_{\rm DK,HA}}{M_{\rm CL,CCPC}}$$
(13)

The recovery rate of waste energy can be divided into two aspects. The recovery rate of waste gas energy  $V_{2,2}$  and solid energy  $V_{2,3}$ , which are the vital order parameters that have significant effect on energy saving, the equation are:

$$V_{2,2} = \frac{E_{\rm RK,FG} + E_{\rm IC,FG} \times 80\%}{E_{\rm DK,FG} + E_{\rm RK,FG} + E_{\rm IC,FG} + E_{\rm CL,FG}} 100$$
(14)

$$V_{2,3} = \frac{E_{\rm IC,RGPC} - E_{\rm RGPC,DHL} + E_{\rm DK,DGPC} - E_{\rm DGPC,DHL}}{E_{\rm IC,RGPC} + E_{\rm DK,DGPC}} 100$$
(15)

Owning to its great thermal content, the flue gas from the incinerator is returned to the waste heat recovery boiler for energy recovery. Following the previous study, the overall heat transfer efficiency of the waste heat recovery boiler is assumed as 80% [27].

The values of the above order parameters were evaluated using eqs. (7)-(15). Most of the order parameters have a positive effect on promoting the orderly development of the calcination system, except for  $V_{1,2}$  and  $V_{2,1}$ . In addition, based on the same CCPC production capacity, the corresponding parameter values at the upper and lower limit operating conditions are denoted as  $V_{m,n}^{ul}$  and  $V_{m,n}^{ll}$ . The SD and all intermediate data were also evaluated considering all the operating conditions remain unchanged are shown in tab. 3.

	V1.1	V1.2	V1.3	V1.4	V1.5	V <sub>2.1</sub>	V2.2	V2.3
$V_{\mathrm{m,n}}$	70.007	5	81.372	4.178	82	0.910	82.409	26.046
$V_{m,n}^{\mathrm{ul}}$	71.329	8	100	4.231	85	1.318	100	100
$V_{m,n}^{11}$	66.795	4	20.202	4.126	78	0.442	17.239	26.046
$EC_{m}(V_{m,n})$	0.709	0.750	0.767	0.499	0.57	0.465	0.787	0.000
Senm,n	0.222	0.196	0.185	0.316	0.291	0.324	0.171	0.000
$\omega_{m,n}$	0.205	0.212	0.215	0.181	0.187	0.270	0.331	0.399
$OD_{\rm m}(IG_{\rm m})$	0.666					0.386		
SD	0.507							

Table 3. The calculated SD of calcination process

## Discussion

### Resource utilization

Figure 2(a) shows the Sankey diagram of the material flow in the drying kiln. As depicted, the percentage of GPC mass going into the drying kiln is of 62.105%, which is then decreased by 6.537% due to moisture evaporation, leaving the kiln as the flue gas eventually. Similarly, fig. 2(b) shows the Sankey diagram of material flow in the rotary kiln. As depicted, the dried GPC accounts for 45.744% of the material flow that later output as CPC. A decrease of the mass of 11.136% is recorded after the calcination in the rotary kiln. This is attributed to the un-burnt dried GPC to incinerator, consumption due to combustion of carbon, sulfur, and volatile as well as moisture evaporation. Meanwhile, the mass percentage of flue gas is of 63.105% of the mass output. The large portion of flue gas is generated by the various combustion reactions of materials and original air supply to the rotary kiln.

The Sankey diagram of material flow in the incinerator is shown in fig. 4(c). One can observe that the mass portion of un-burnt dried GPC together with the flue gas from the rotary kiln to the incinerator account for 33.889% of the total mass. After the secondary combustion, the mass percentage of flue gas raises significantly up to 99.023%, indicating almost all materials have been completely consumed by combustion. Finally, fig. 2(d) shows the Sankey diagram of material flow in the cooler. In the cooler, the mass portion of the CPC only accounts for 7.149%, while the majority of mass is accounted for the circulating cooling water, *i.e.* 88.131%. With a close circulating system, there is no mass loss for the cooling water passing through the cooler, *i.e.* only 3.574%. The sprayed water converted into flue gas; which in turn accounts for 39.768% of the mass flow outputs excluding the mass of the circulating cooling water.

In terms of resource utilization, two major aspects are considered in the present study. The first one focuses on the waste recovery strategy. The other one focuses on the reduction of resource consumption. Firstly, the waste recovery consists of solid and gas waste energy recovery with several different recovery principles. Referring to fig. 2(c), it can be observed that the mass portion of returned GPC and dust contribute 0.802% and 0.175% of the total mass, respectively. The returned GPC can be sent back to the rotary kiln, and dust can be collected by the dust collector. Similarly, in the cooler, fig. 2(d), a small portion of dust (*i.e.* 0.005%) can also be collected by the dust collector. Thus, the solid waste produced in the cal-

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Figure 2. Sankey diagram of material flow in the four different equipment; (a) Sankey diagram of the material flow in the drying kiln, (b) Sankey diagram of the material flow in the rotary kiln, (c) Sankey diagram of the material flow in the incinerator, and (d) Sankey diagram of the material flow in the cooler

cination process has been fully utilized. Although flue gas is generated in each equipment, figs. 4(a)-4(d), only flue gas from the rotary kiln and the incinerator is utilized. As the recovery rate of waste gas is of 81.372%, it is worth to develop some strategies for fully utilizing the waste gas. Subsequently, aiming to reduce the resource consumption, it is necessary to develop an effective measure to reduce the carbon loss, *i.e.* 5%, during the calcination process using rotary kiln in the current system operation.

## Energy saving

Figure 3(a) shows the Sankey diagram of the energy flow in the drying kiln. As depicted, physical sensible heat of the hot air is the main heat source of the drying kiln, corresponding to 95.938% of the energy input. Meanwhile, as the main function of the drying kiln,

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Figure 3. Sankey diagram of energy flow in the four different equipment; (a) Sankey diagram of the energy flow in the drying kiln, (b) Sankey diagram of the energy flow in the rotary kiln, (c) Sankey diagram of the energy flow in the incinerator, and (d) Sankey diagram of the energy flow in the cooler

majority of energy output is contributed by moisture evaporation heat, *i.e.* 63.574%. Nonetheless, the energy loss due to the unavoidable flue gas physical sensible heat accounts up to 21.095%. Although the energy loss caused by the flue gas is considerably high, due to its relatively low temperature, *i.e.* 99.5 °C, and unstable generation, it is considered uneconomical to recovery the heat from the flue gas. On the other hand, the physical sensible heat of the dried GPC accounts for 13.057% of the energy input. This energy portion could be fully utilized instead of dissipating to surroundings during the transportation process. Figure 3(b) shows the Sankey diagram of the energy flow in the rotary kiln. As the main heat source supporting the normal operation of the rotary kiln, the combined heat combustion of the volatile and the carbon account for 90.760% of the energy input. Furthermore, natural gas combustion heat also contributes 6.430% of the energy input. Based on the Sankey diagram, one can notice that the contribution of the natural gas combustion could be reduced replacing by the available heat in the dried GPC.

In terms of energy output, physical sensible heat of CPC and flue gas account for 38.159% and 33.493%, respectively. In addition, the heat dissipation of the rotary kiln accounts for 23.437% of the energy output. Due to structural restriction of rotary kiln design, it is impractical to reduce the energy loss by increasing the surface insulation. Therefore, although the surface temperature of the rotary kiln could reach around 250 °C during the calcinations process, there is limited choice could be applied to reduce the heat loss through the surface.

The Sankey diagram of energy flow in the incinerator is shown in fig. 3(c). From the figure, it can be observed that majority of the energy input is contributed by the heat of combustion of volatile, sulfur and carbon in un-drop down GPC to incinerator and combining a total of 76.984% of the energy input. Together with the heat of flue gas coming from rotary kiln, it makes up to 98.860% of the heat source. The heating process occurring in the incinerator fully utilizes the thermal potential of rotary kiln flue gas, and the heat of incinerator flue gas, accounting for 98.658% of the heat output. Part of the heat output will be utilized for the downstream heat recovery system. Figure 3(d) shows the Sankey diagram of the energy flow in the cooler. Majority of the energy input is attributed to the CPC physical sensible heat, *i.e.* 59.028%, coming from the rotary kiln. To maintain the CPC temperature to be below 80 °C for transportation, the cooling technology combing internal water spraying and external water circulating is adopted in the cooler. Subsequently, spray water vaporization heat is the majority heat output of the cooler, *i.e.* 41.326%. In addition, the net heat removal of external circulating water accounts for 15.712%.

For energy saving, two main strategies are commonly considered: energy recovery and energy consumption reduction. Firstly, energy recovery can be achieved with solid waste and gas waste heat recovery. With distinguished characteristics of solid and gas waster, significantly different recovery methods were adopted. For the solid waste heat, the energy loss of material between each equipment with little temperature drop during the transportation is negligible. However, due to the huge temperature difference between the dried GPC from drying kiln, 80 °C, to rotary kiln, 25 °C, shown in tab. 2, the dried GPC physical sensible heat accounting for 13.057% of the energy output in fig. 3(a) is necessary to find recycling ways. Similarly, the dissipation heat loss of returned GPC from the incinerator to the rotary kiln is worthwhile for recycling. For the gas waste heat, excluding the utilized flue gas heat in the rotary kiln and incinerator, the flue gas heat in the drying kiln and the cooler is coarsely wasted. As it is uneconomical to recovery the flue gas heat in drying kiln due to its low temperature and unsteady generation, developing a strategy to recycle the flue gas heat in the cooler is imperative.

One the other hand, from the aspect of energy consumption reduction, as the highest energy consumption equipment in the process, it is crucial to optimize the energy utilization in the drying kiln. As shown in fig. 3(b), natural gas combustion heat accounts for 6.430% of the energy input which could be optimized by improving the process operation system or alternating the equipment design. Furthermore, the cooling method could also be optimized to improve the heat exchange efficiency of the external circulating water.

#### Possible improvements

The synergy of the material flow, Sub<sub>1</sub>, and energy flow, Sub<sub>2</sub>, has a significant impact on the performance of the calcination process. The random fluctuation of each order parameter overwhelms the normal manufacturing operation, disturbing the material and energy flow. Therefore, the order degree of each subsystem indicates the SD of the whole calcination process. According to the current calcination process, the SD is 0.507 as summarized in tab. 3. Similarly, the order degree of Sub<sub>1</sub> and Sub<sub>2</sub> are of 0.666 and 0.386, respectively. The order degrees reflect that essential improvement should be taken to adjust the order parameters. From the standard weight of efficacy coefficient in Sub<sub>1</sub>, the recovery rate of waste gas,  $V_{1,3}$ , is found significantly higher than the other four. This is attributed to the inefficacy use of flue gas in the drying kiln and the cooler.

Furthermore, it is also desired to develop some feasible plans to reduce the carbon loss,  $V_{1,2}$ , due to its higher standard weight. In Sub<sub>2</sub>, the recovery rate of the waste gas energy,  $V_{2,2}$ , and the recovery rate of solid waste energy,  $V_{2,2}$ , are the dominant factors affecting the order degree of the subsystems. Hence, it is essential to develop an effective measure to recovery flue gas heat from the drying kiln and cooler, and minimizes the heat dissipation of the dried GPC and the returned GPC during the transportation process.



Figure 4. Sankey diagram of the material flow with the suggested improvements



Figure 5. Sankey diagram of the energy flow with the suggested improvement

In order to improve the resource utilization, energy-saving and synergy performance of the calcination process, several suggestions are proposed in the present study. Firstly, as shown in fig. 1, a burner set at the head of rotary kiln is used to maintain a higher calcination temperature and ensure the product quality by adding fuel for supplementary combustion. This operation concept has been successfully applied in many anode plants in China. None-theless, some problems have also been exposed, including high carbon loss and incomplete combustion of volatile caused by the calcination location close to the head of rotary kiln. Therefore, it is suggested to install electrical fans at the rotary kiln shell to enhance the combustion completeness inside the rotary kiln [28]. As a result, the combustion heat of volatile in the dried GPC could be fully utilized instead of consuming external fuel. Meanwhile, it can also reduce the temperature at the rotary kiln head, minimizing the carbon loss during the calcination process.

Secondly, the temperature of dried GPC from drying is 80 °C, and the temperature of GPC returned from incinerator is 200 °C. Therefore, it is better ro recovery the heat of the two materials. Blending bins between drying kiln and rotary kiln should take heat preservation to reduce temperature drop of dried GPC and returned GPC inside. On the other hand, the transportation path of the returned GPC from the incinerator to the rotary kiln could be optimized for a more direct delivery to minimize heat loss.

Thirdly, with the high heat value of the cooler flue gas, it is recommended to utilize the waste heat by leading it to the incinerator for secondary heating and further utilize in the waste heat recovery boiler system. Last but not least, as the cooling method of spraying water inside is more efficient comparing to the circulating water outside, it is suggested to adopt the internal water spraying completely for cooling in the cooler. The external cooling water circulation can only be employed as a standby operation measure.

Based on the previous discussion, considering all improvement measures are implemented, the SD of the whole calcination process is then recalculated. Figures 4 and 5 show the resultant mass and energy balance of the whole calcination process with suggested improving measures. Obviously, the SD is raised up to 0.809. This encouraging result is caused by the lower carbon loss, more effective utilization of waste gas and solid, and less external energy consumption. With the improvement, a more orderly calcination system between material and energy flow could be obtained. The corresponding improved indices are summarized in tab. 4.

	$V_{1.1}$	$V_{1.2}$	$V_{1.3}$	$V_{1.4}$	$V_{1.5}$	$V_{2.1}$	$V_{2.2}$	V <sub>2.3</sub>
$V_{\mathrm{m,n}}$	70.763	4	88.188	4.179	82	0.577	82.997	100
$V_{m,n}^{\mathrm{ul}}$	71.329	8	100	4.231	85	1.651	100	100
$V_{m,n}^{11}$	69.884	4	20.461	4.127	78	0.404	18.375	26.046
$EC_{m}(V_{m,n})$	0.609	1.000	0.851	0.500	0.571	0.861	0.792	1.000
S <sub>enm,n</sub>	0.275	0.000	0.125	0.315	0.291	0.117	0.168	0.000
ωm,n	0.182	0.250	0.219	0.171	0.178	0.325	0.307	0.368
$OD_{\rm m}(IG_{\rm m})$	0.735					0.891		
SD	0.809							

Table 4. The recalculation of SD of improved calcination process

#### Conclusions

- The present paper provides a comprehensive analysis of the calcination process using a synergy model based on the mass and energy conservations in conjunction with the synergy theory. All four major equipment involved in the process is discussed in detail.
- For the material flow analysis, the product yield of GPC is evaluated as 70.007%. For the energy flow analysis, the overall energy efficiencies of the drying kiln, rotary kiln, incinerator, and the cooler are 63.574%, 37.709%, 76.782%, and 74.758%, respectively.
- In order to evaluate the system status, order parameters tailored for the calcination system are defined based on the synergy model. The value of SD is evaluated as 0.507, indicating a relatively low degree of cooperation between the material flow and energy flow.
- Several possible improvements are proposed to improve the calcination process on resource utilization and energy saving. Electrical fans are recommended to be installed at the rotary kiln shell of the rotary kiln shell and adjust the process of the operating system. Moreover, it is proposed to fully recover the waste heat of the dried GPC, the returned GPC and flue gas from the cooler. Furthermore, a direct cooling method using internal water spray is recommended to be fully implemented in the cooler. With the suggested improvements, the SD is recorded having a substantial enhancement up to 0.809, indicating a more orderly calcination process within the system.
- Based on our previous work, the present study extended the model considerations and advanced the formulations for the synergy analysis. The synergy analysis also draws new findings and conclusions on the calcination process. With the advanced mathematical model and considerations of new order parameters, it provides new sights and a more comprehensive energy-saving route for other researchers.

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