

Thermodynamic and Environmental Impact Assessment of Calcium Oxide Formation in Clinker Production

Boyaghchi, F. A.

Faculty of Engineering, Alzahra University, Vanak, Tehran, Iran

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ABSTRACT: In this research, energetic and exergetic analysis of calcium oxide formation, CO₂ emissions, and environmental effects during the clinker production process in rotary kiln are investigated. First the energy and exergy analysis of rotary kiln were carried out according to the first and second law of thermodynamics based on dead state, respectively. Consequently, the amount of CO₂ emissions associated with calcination and fuel combustion processes, improvement potential and sustainability index are determined. In this system, the energy efficiency is calculated to be higher than the exergy efficiency, e.g. 53.4% and 28.6% for the energy and exergy efficiencies. The exergy destruction due to irreversible calcination and fuel combustion processes is determined 33,884 kW which is 40.6% of inlet exergy. Results also show that 18.4% of exergy input is lost by exhaust hot gases. The total emissions are estimated 157,228 kg/hr, which 31.3 % of it is due to calcination and 0.9 % of it is due to fuel combustion. The improvement potential (IP) and sustainability index (SI) are calculated 24,193.176 kW and 1.4, respectively.

Key words: Energy, Exergy, Efficiency, Greenhouse Gas

INTRODUCTION

Calcium oxide (CaO), commonly known as lime or quicklime, is an energy intensive product characterized by high CO₂ emissions, and it contains more than 67% of clinker in rotary kiln. During this process a lot of CO₂ emits during the decomposition of CaCO₃. Quicklime production accounts for about 8% of total CO₂ emissions from all human activities (IEA, 1999). The main energy consumption of lime production is located in the calcination process and accounts for more than 90% of the total energy consumption. The calcination process also accounts for 99% of the impact of lime production on global warming (Sagastume *et al.*, 2012). Moreover, global CO₂ emissions may be increased by 50% more than 1997 level. This accumulation will lead to increase of global mean temperature (GMT) in 21st century up to 5.8°C as high as current temperature if this level of emissions continues (Mahmoud *et al.*, 2009; Dias & Arroja, 2012; Shafiepour Motlagh and Farsiabi, 2007; Ataei *et al.*, 2009; Xing *et al.*, 2010; Montero Lorenzo *et al.*, 2011; Cui *et al.*, 2011; Quesada-Rubio *et al.*, 2011; Wang *et al.*, 2011; Nava-Martinez *et al.*, 2011; Roshan *et al.*, 2012; Hashemian *et al.*, 2013). Fuel combustion is also associated to CO₂ emission in the calcination process. This situation causes lime one of the industrial products with the highest emission of CO₂ associated to its production. As the quantity of

CO₂ emitted due to the dissociation of CaCO₃ is constant, the final amount of CO₂ emitted depends on the efficiency of the fuel consumption in the kiln (Hoening & Schneider, 2002; Wolter & Fuchs, 2007).

The evaluation of the thermal efficiency of quicklime kilns has been the subject of several studies (Ochoa *et al.*, 2010; Boynton, 1980; Sagastume & Vandecasteele, 2011). The second law of thermodynamics is instrumental in providing insights into environmental impact. The most appropriate link between the second law and environmental impact has been suggested to be exergy, in part because it is a measure of the departure of the state of a system from that of the environment (Kanoglu *et al.*, 2012). Moreover, the main purpose of exergy analysis is to detect and assess quantitatively the thermodynamic imperfections' causes of thermal and chemical processes. The exergy method of thermodynamic analysis is based upon both the first and the second laws of thermodynamics together, while the energy analysis is based upon the first law only. It is a feature of the exergy concept to allow quantitative assessment of energy degradation (Morris & Szargut, 1986).

Exergy analysis is consequently linked to sustainability because in increasing the sustainability of energy use, we must be concerned not only with

*Corresponding author E-mail: fahmadi@alzahra.ac.ir

loss of energy, but also loss of energy quality (or exergy). A key advantage of exergy analysis over energy analysis is that the exergy content of a process stream is a better valuation of the stream than the energy content, because the exergy indicates the fraction of energy that is likely useful and thus utilizable. This observation applies equally at the component level, the process level, and the life cycle level. Application of exergy analysis to a component, process, or sector can lead to insights regarding how to improve the sustainability of the activities comprising the system by reducing exergy losses (Kanoglu *et al.*, 2012).

In this respect, the energy and exergy has been analyzed in a rotary burner with pre-calcinations in cement production in several studies (Camdali *et al.* 2004; Koroneos *et al.* 2005; Utlü, 2006; Sogut *et al.*, 2009; Ari, 2011; Sax; Madloul *et al.*, 20012). A dry type rotary kiln system was investigated with a kiln capacity of 600 ton clinker per day (Engin & Ari, 2004). They found that about 40% of the total input energy was lost through hot flue gas, cooler stack and kiln shell. In this work, the energy, exergy, environmental impact analysis, energetic and exergetic efficiency, Improvement potential and sustainability index for quicklime formation in rotary kiln are investigated.

MATERIALS & METHODS

The energy and exergy analysis are applied for evaluation of the rotary kiln performance. The mass, energy and exergy balances of the rotary kiln are therefore established. The system is operating under steady state conditions. The kinetic and potential energy of the stone and gas flows are negligible compared with their enthalpy. For a control volume operating under steady state conditions the mass balance given by

$$\sum \dot{m}_{in} = \sum \dot{m}_{out} \quad (1)$$

where \dot{m}_{in} and \dot{m}_{out} indicate input and output flow rates, respectively.

The general steady-state energy balance can be expressed as:

$$\sum \dot{E}_{in} = \sum \dot{E}_{out} \quad (2)$$

where \dot{E}_{in} and \dot{E}_{out} indicate the input and output energy rates of the rotary kiln. According to the first law of thermodynamics the energy of flow is (Çengel, & Boles, 2006):

$$\dot{E} = \dot{m}c_p(T_2 - T_1) \quad (3)$$

Where c_p and T are the specific heat and temperature of flow, respectively. The first law efficiency of the rotary kiln is calculated as (Gutiérrez *et al.*, 2012):

$$\eta = \frac{H_R (\text{CaOwt}\%)}{\dot{E}_F} \quad (4)$$

where H_R is the reaction enthalpy, obtained by calculating the formation and sensitive enthalpies of products (CaO and CO₂) and reactant (CaCO₃), and (CaOwt%) is the percentage of CaO in the lime. The energy supplied by the fuel, \dot{E}_F , is calculated as:

$$\dot{E}_F = \dot{m}_F \frac{\sum y_i (\text{LHV})_i}{\sum y_i M_i} \quad (5)$$

where y_i and LHV_i are molar fraction, molar lower heating value and molar mass of i th component of fuel (Table 1), respectively.

Exergy can be described as the maximum available work which can be produced by a system when it comes to equilibrium with a reference environment. It has been showed that an exergy analysis is a powerful tool in the thermodynamic analyses of energy systems. It is employed to evaluate quantitatively the causes of thermodynamic imperfection of the process under

Table 1. Natural gas compositions, - COMGAS

Component	Volume (%)	Mass (%)	$\overline{\text{LHV}}$ (kJ/kmol)	\overline{ex} (kJ/kmol)
CH ₄	89.92	80.92	802,000	831,650
C ₂ H ₆	13.03	13.64	1,429,000	1,495,840
C ₃ H ₈	0.78	1.94	2,045,000	2,154,000
C ₄ H ₁₀	0.07	0.23	2,660,000	2,805,800
C ₅ H ₁₂	0.01	0.04	3,276,000	3,463,300
CO ₂	0.48	1.20	-	19,870
N ₂	1.28	2.03	-	720

consideration. In addition, the exergy analysis usually aims to determine the maximum performance of the system and identify the equipment in which exergy loss occurs, and indicates the possibilities of thermodynamic improvement of the process under consideration (Ari, 2011).

The exergy balance of a thermal system is given by

$$\sum \dot{E}x_{in} = \sum \dot{E}x_{out} + \dot{E}x_D \quad (6)$$

where $\dot{E}x_{in}$ is the exergy rate of the input flows, $\dot{E}x_{out}$ is the exergy rate of the output flows and $\dot{E}x_D$ is the exergy destroyed in the process.

In the absence of electricity, magnetism, surface tension and nuclear reaction the exergy content of a flow equals;

$$\dot{E}x_T = \dot{E}x_{ph} + \dot{E}x_{ch} \quad (7)$$

where $\dot{E}x_{ph}$ is the physical exergy rate and $\dot{E}x_{ch}$ is the chemical exergy rate. The physical exergy rate is obtained (Çengel, & Boles, 2006):

$$\dot{E}x_{ph} = \dot{m}[(h - h_o) - T_o \cdot (s - s_o)] \quad (8)$$

where h is the specific enthalpy and s the specific entropy, the subscript o refers to the reference environment.

The enthalpy change, according to the first principle of thermodynamics, is given by (Çengel, & Boles, 2006):

$$(h - h_o) = c_p(T - T_o) \quad (9)$$

Combining the first and the second principle of thermodynamics, the entropy change is given by (Çengel, & Boles, 2006):

$$s - s_o = c_p \ln\left(\frac{T}{T_o}\right) - R \ln\left(\frac{P}{P_o}\right) \quad (10)$$

The pressure term is applied to calculate the entropy of gases, not for liquids and solids (Çengel, & Boles, 2006):

The specific physical and chemical exergy of a gas mixture are given by (Çengel, & Boles, 2006):

$$\dot{E}x_{ph} = \frac{\dot{m}}{M} \sum_i y_i \bar{e}x_{ph,i} \quad (11)$$

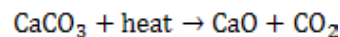
$$\dot{E}x_{ch} = \frac{\dot{m}}{M} \left[\sum_i y_i \bar{e}x_{ch,i} + RT_o \sum_i y_i \ln(y_i) \right] \quad (12)$$

Where $\bar{e}x_{ph}$ and $\bar{e}x_{ch}$ are the standard molar chemical exergy and M is molecular weight of gas mixture.

The chemical exergy of substances in reaction $M + N \rightarrow P$ is related to the Gibbs free energy of the reaction ΔG_o by the equation (Bejan *et al.*, 1996):

$$\dot{E}x_{ch,M} + \dot{E}x_{ch,N} - \dot{E}x_{ch,P} = \Delta G_o \quad (13)$$

In this research the chemical reaction can be written as follow:



The chemical exergy of substances is tabulated (Gutiérrez *et al.*, 2012). The exergy of the heat is given by (Çengel, & Boles, 2006):

$$\dot{E}x_Q = \left(1 - \frac{T_o}{T}\right) \dot{Q} \quad (14)$$

where \dot{Q} is the rate of heat loss through walls, calculated from energy balance and T is the rotary kiln temperature.

The exergy efficiency of lime production is expressed as (Gutiérrez *et al.*, 2012):

$$\epsilon = \frac{\dot{E}x_{ch,Cac}}{\dot{E}x_{ch,f}} \quad (15)$$

where $\dot{E}x_{ch,Cac}$ is the chemical exergy rate of lime, $\dot{E}x_{ch,f}$ is the fuel chemical exergy which can be calculated

$$\text{with: } \dot{E}x_{ch,f} = \dot{m}_f \frac{\sum y_i \bar{e}x_i^{ch}}{\sum y_i M_i} \quad (16)$$

where y_i , $\bar{e}x_i^{ch}$ and M_i are molar fraction, molar chemical exergy and molar mass of i th component of fuel, respectively (Table 1).

The mass of exit gases equals (Gutiérrez *et al.*, 2012):

$$m_g = m_F + m_a + m_{CO_2} \quad (17)$$

where the last term on the right hand side is the CO_2 released during calcination.

The sustainability of the fuel resource is expressed by a sustainability index (SI) as the inverse of the depletion number (Kanoglu *et al.*, 2012):

$$SI = \frac{1}{D_p} \quad (18)$$

The relationship between the depletion factor and the exergy efficiency is

$$\epsilon = 1 - D_p \quad (19)$$

The maximum improvement in the exergy efficiency for a process can be obtained when the exergy loss or irreversibility $\dot{E}x_{in} - \dot{E}x_{out}$ is minimized (Van Gool, 1997). Consequently, he suggested "improvement potential", denoted IP which describes recovery potential in exergy losses or irreversibility's of the system as following:

$$IP = (1 - \epsilon)(\sum \dot{E}x_{in} - \dot{E}x_{out}) \quad (20)$$

RESULTS & DISCUSSION

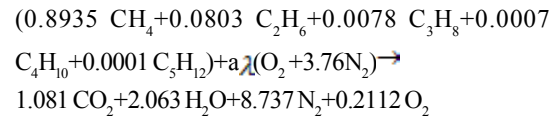
Fig. 1 indicates the rotary kiln included the input flows of fuel, air, and limestone and the output flows of lime and exhaust gases. It works 24 h a day without interruptions. The mass balance of the rotary kiln can be written by:

$$\dot{m}_{Ls} + \dot{m}_F + \dot{m}_a = \dot{m}_l + \dot{m}_g \quad (21)$$

where \dot{m}_{Ls} , \dot{m}_F , \dot{m}_a , \dot{m}_l and \dot{m}_g show the mass rate of limestone, fuel, air, lime and exhaust gases, respectively. In equation 18, the mass rate of exhaust gases (\dot{m}_g) equals the sum of the mass rate of combustion gases ($\dot{m}_F + \dot{m}_a$) and the mass of CO_2 obtained from calcinations (Gutiérrez *et al.*, 2012).

The mass balance for rotary kiln is listed in Table 2.

According to Table 1, combustion process with 10% excess air can be modeled as:



The mass flow rate of products during combustion process can be estimated by (Khartchenko, 1998):

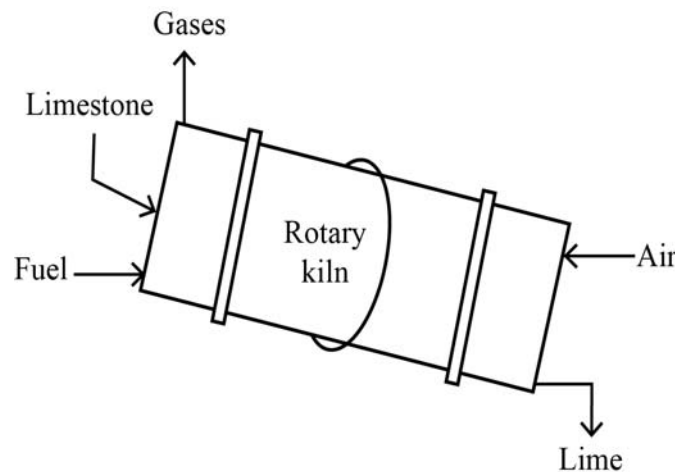


Fig. 1. Rotary kiln

Table 2. Mass Balance

Input Flows (kg/hr)	
Limestone	120,000
Air	97,509
Fuel	5,320
Total	222,829
Output Flows (kg/hr)	
Lime	70,833
Gases	151,996
Total	222,829

$$\dot{m}_{\text{CO}_2, \text{total}} = (\dot{m}_{\text{CO}_2})_f + (\dot{m}_{\text{CO}_2})_{\text{sto}} \quad (22)$$

$$\dot{m}_{\text{H}_2\text{O}, \text{total}} = (\dot{m}_{\text{H}_2\text{O}})_{\text{sto}} + 1.6 \dot{m}_f \omega A F_{\text{act}, d} \quad (23)$$

$$\dot{m}_{\text{N}_2, \text{total}} = (\dot{m}_{\text{N}_2})_f + 0.79 \dot{m}_f A F_{\text{act}, d} \quad (24)$$

$$\dot{m}_{\text{O}_2} = 0.21 \dot{m}_f (\lambda - 1) A F_{\text{sto}, d} \quad (25)$$

Where $(\dot{m}_{\text{CO}_2})_f$ and $(\dot{m}_{\text{N}_2})_f$ are the mass flow rates of CO_2 and N_2 existing in fuel. $(\dot{m}_{\text{CO}_2})_{\text{sto}}$ and $(\dot{m}_{\text{H}_2\text{O}})_{\text{sto}}$ are the mass flow rates of CO_2 and H_2O release during stoichiometry combustion process. ω and λ are the humidity relative ($0.01 \text{ kg}_{\text{H}_2\text{O}}/\text{kg}_{\text{dry air}}$) and excess air (10%), respectively. $A F_{\text{act}, d}$ is actual air-fuel mass ratio in dry air which is obtained as:

$$A F_{\text{act}, d} = \lambda A F_{\text{sto}, d} = 4.76 \frac{\rho_a}{\rho_g} \left(n_c + \frac{1}{2} n_H - n_o \right) \quad (26)$$

In above formula is the air density (1.293 kg/m^3), is the gases density (0.790 kg/m^3) and $n_c (=1.081)$, $n_H (=2.063)$ and $n_o (=0.0048)$ are carbon, hydrogen and oxygen molar amounts of fuel obtained from Table 1. Table 3 shows emissions from rotary kiln during these process. It is obvious that the total emissions due to fuel combustion is $108,561 \text{ kg/hr}$, which $63,510 \text{ kg/hr}$ of it is CO_2 emissions.

Fig. 2 shows the percentage of emissions during the calcination and combustion processes. It is obtained that 40.3% of total emissions is associated to CO_2 emissions.

Table 4 indicates energy balance of input flows and output flows for rotary kiln which are calculated using equation 27.

$$\dot{E}_{\text{Ls}} + \dot{E}_{\text{F}} + \dot{E}_{\text{a}} = \dot{E}_{\text{l}} + \dot{E}_{\text{g}} + \dot{E}_{\text{R}} + \dot{E}_{\text{loss}} \quad (27)$$

where \dot{E}_{Ls} , \dot{E}_{F} , \dot{E}_{a} , \dot{E}_{l} , \dot{E}_{g} and \dot{E}_{loss} are energy rates of limestone, fuel, air, lime, gases and losses, respectively.

Table 3. Environmental impact

Reactants (kg/hr)				
Fuel	O ₂	N ₂	Total	
5,320	27,314	75,927	108,561	
Products (kg/hr)				
CO ₂	H ₂ O	N ₂	O ₂	Total
14,343	16,458	75,927	1,833	108,561
49,167				157,728
Total CO ₂			63,510	

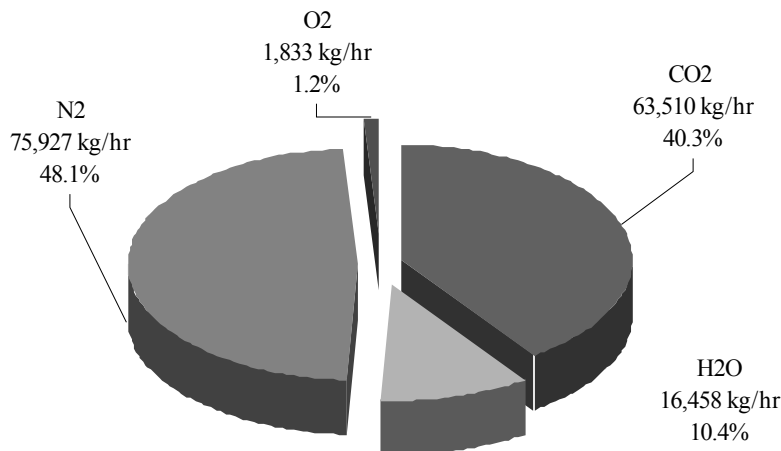


Fig. 2. Total emissions. Given as the percentages of total emissions (157,728 kg/hr)

Table 4. Energy Balance

Input Flows		
	T (K)	\dot{E} (kW)
Limestone	298	0
Air	298	0
Fuel	298	70,883
Total	-	70,883
Output Flows		
	T (K)	\dot{E} (kW)
Lime	1248	8,336
Gases	1373	20,317
Enthalpy of reaction	-	39,497
Losses	-	2,736
Total		70,883
Thermal efficiency = 53.4%		

The energy of gases is calculated as:

$$\dot{E}_g = \dot{m}_g \frac{\sum y_i \bar{c}_{p,i} \Delta T}{\sum y_i M_i} \quad (i = \text{CO}_2, \text{H}_2\text{O}, \text{N}_2, \text{O}_2) \quad (28)$$

where \dot{m}_g , y_i , $\bar{c}_{p,i}$, M and ΔT are exhaust gas mass flow, molar fraction, molar specific heat, molar mass and temperature differences after combustion, respectively.

Fig. 3 shows the energy input flows of limestone, fuel and air; the energy output flows of lime and exhaust gases, enthalpy of calcination and the heat loss through the walls of the rotary kiln. The thermal efficiency of rotary kiln is 53.4% obtained using equation 4.

The process for which the exergy analysis performed is assumed to be an open system under the

steady-state working conditions. Thereference pressure and temperature for the dead condition are assumed to be 295 K and 101 kPa, respectively.

There are both physical (thermo-mechanical) and physical exergies in the process since chemical reaction takes place in rotary kiln. The temperature in which the decomposition of CaCO_3 into CaO and CO_2 takes place is assumed 900°C (Ari, 2011).

Table 5 shows the exergy balance of rotary kiln calculated using equations 6 to 12, 14 and 16. Exergy destruction of rotary kiln is calculated by equation 6.

Fig. 4 illustrates the exergy of input and output flows in rotary kiln. The exergy efficiency of rotary kiln is calculated using equation 15.

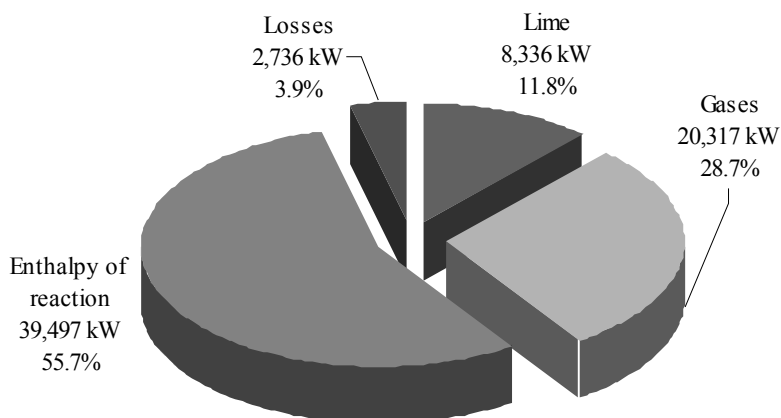


Fig. 3. Energy diagram. Given as the percentages of energy input (70,883kW)

Table 5. Exergy Balance

Input Flows				
	$T(K)$	$\dot{E}x_{ph}(kW)$	$\dot{E}x_{ch}(kW)$	$\dot{E}x_T(kW)$
Limestone	298	0	6,006	6,006
Air	298	0	1,282	1,282
Fuel	298	0	76,137	76,137
Total	-	0	83,425	83,425
Output Flows				
	$T(K)$	$\dot{E}x_{ph}(kW)$	$\dot{E}x_{ch}(kW)$	$\dot{E}x_T(kW)$
Quicklime	1248	9,180	23,863	33,043
Gases	1373	33,623	262	15,345
Loss	-	1,153	0	1,153
Total	-	-	-	49,541
$\dot{E}x_D = 83,425 - 49,541 = 33,884 \text{ (kW)}$				
Exergy efficiency=28.6%				
SI=1.4				
IP=24,193.176 kW				

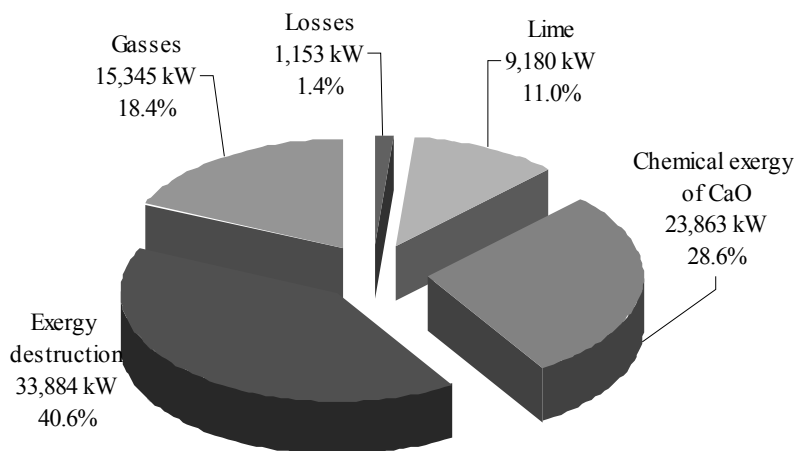


Fig. 4. Exergy diagram. Given as the percentages of exergy input (83,425 kW)

CONCLUSIONS

This study is carried out the energy, exergy and environment analysis in rotary kiln. The following outcomes were achieved:

- The maximum energy loss is found in the exhaust gases where 28.7% of the input energy was lost to the environment. 3.9 % of the energy loss was identified for wall losses while 11.8% is associated with lime.
- The total energy efficiency of cement rotary kiln is 53.4% based on inlet fuel energy.
- In terms of exergy destruction, the major loss is observed in the combustion and chemical reaction in kiln with the value of 40.6% of the exergy input. 18.4%, 11.0% and 1.4% of exergy input were lost by exhaust gases, lime and wall losses, respectively.
- The total exergy efficiency of cement rotary kiln is 28.6% based on the inlet exergy.
- The total emissions are 157,228 kg/hr, which 40.3% of it is CO₂.

- The amount of CO₂ emission during calcination and combustion is calculated 63,510kg/hr which 49,167 kg/hr of it is produced during calcination.
- The improvement potential (IP) and sustainability index (SI) are calculated 24,193.176 kW and 1.4, respectively.

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