P. C. Okonkwo, S. S Adefila, A.S Ahmed/ International Journal of Engineering Research and Applications (IJERA) ISSN: 2248-9622 www.ijera.com
Vol. 2, Issue 2,Mar-Apr 2012, pp.616-628

Development of Process Simulation Model for Lime Production

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Abstract

Lime production is a process that entail complex material and energy transport and chemical reaction. Efficient lime production processes are realized via well structured design models approach . Simulation of such process design models essentially provides Information on the attractive features and technical limitations of such processes which serves as guide for normal operations. The choice of appropriate descriptive models forms the basis of a valuable simulation of the process of limestone calcination. This work aim at developing a systematic development of process simulation models for lime production via calcinations of limestones . The various transport phenomena and chemical reaction models prevailing in the conversion of limestone to lime in the kiln are presented in a fundamental and systematic manner.

The calcination models which forms a layer model for overall kiln design have been simulated over a temperature range of 900 to 1080'C at varying pressures for Jakura and Ukpilla limestone sources found in the central region of Nigeria. The results show the variation of calculation times in respect of limestone particle size and the kiln pressures. Employment of the models in the simulation of lime production will ensure high utility of the simulation tool.

Keywords ; Lime, Calcination, Transport phenomena, Simulation, Process model, Design

1.0 Introduction

The operations of a lime kiln can be greatly simplified by the use of simulation results obtained in the process of conception and design. The origin of the deign of kilns can be traced back to the Egyptian civilization. Lime productions however have developed from the primitive batch kilns to the modern sophisticated continuous kilns ii the last 50 years [1] . The dependency of the kiln operations on the key parameters is usually evaluated with a typical kiln simulation model. The kiln is the heart of lime production process and the efficiency of the entire process of lime production will be dependent on the efficiency of the kiln hence its design.

A typical approach to obtaining the complete process model involves the considerations of the following:

(a) Transport phenomena models
(b) Calcination model
(c) Reactor model

The individual models constitute layer models which are usually integrated to yield the complete kiln model; which can then be simulated over ranges of conditions obtainable in typical operations. Also the layer models are simulated in order to determine the utility and their effect on the overall model. In this work the calcination model of Jakura and Ukpilla limestone sources were simulated over a temperature and pressure range. The simulation exercise entailed the determination of the relationship between the time for complete calcinatlon of limestone particles of known gradation and extent of calanation achieved for a particular period of calcinatlon at specified temperature and pressure. Previous works and studies carried out on the limestones [2] have shown that the calcination reaction is limited by heat and mass transfer across lie layer. Thus the heat and mass transfer limited reaction model [3] was used to simulate the calcination model.
Also the models of the prevailing material and energy transport and simplifying assumptions are presented to enable simulation of the entire kiln operation.

2.0 Theory

2.1 Types of kiln

Kilns employed for burning lime may be divided into two classes[4] : intermittent kilns and continuous kilns. The latter class in turn may be sub-divided into rotary and shaft kilns. The shaft kilns may again be sub-divided into mixed feed and separate feed kilns. The intermittent kilns are usually known as "pot kilns" from types. They are crude, uneconomical and rarely sued except in some occasions because they are expensive to construct. The continuous shaft kilns may be either in a mixed feed or separate feed. In the former class the fuel comes in contact with the limestone, in the latter the fuel is burned and only the product of combustion come in contact with the limestone. The design and operation of kilns have originated from the days of the ancient Egyptians and has developed into modern process industry. Most of the advances have occurred since 1900 with the greatest progress since 1955[5] . Brief description of each of these types are presented herewith.

2.1.1 Field of pot kilns

These are the first generation of kilns. These were usually constructed crudely of stone often on the side of a hill. They were of low height and had steel plates at the top serving as windbreak. After feeding, a wood fire is ignited in the hearth below and burnt for three or five days. When the fire is extinguished, the lime is drawn, after eating, by hand poling to the hearth below with usually about 25% overburned and same percent under burned, and the balance a passable grade of quicklime. These kilns were operated intermittently and involved extensive manual labour.

2.1.2 Vertical mixed feed kiln

These were an improvement on the pot kilns and featured greater height and capacity. These are largely continuous and lined with refectory bricks. These kilns have improved fuel efficiency and were inexpensive to construct but the ash contaminated the lime since the fuel and lime are intermingled in the kiln and discharged together.

2.1.3 .Vertical kilns

Modern vertical kilns are divided into four distinct zones by imaginary horizontal planes. From the top to bottom in sequence are:

a. Stone storage
b. Preheating zone, designed to heat the stone near dissociation temperature
c. Calcining zone where combustion occurs
d. Cooling and discharge zone usually shaped like an inverted, truncated cone at the bottom of which the lime is discharged.

The various design of vertical kilns are:

a. Traditional shaft types
b. Indirect gas-fire, (producer gas)
c. Large-capacity mixed feed
d. Parallel-flow regenerative
e. Double-inclined
f. Annular (ring)

2.1.4 Indirect Gas-fired kiln

In this type solid fuels are supplied to separate furnace, forming gaseous fuels for combustion. These fireboxes may be installed into to the kiln structure or as adjuncts, separated from the kiln except for ducts and tuyeres. These are also known as producer gas kilns.

Generally, a more even heat results from the injection of the hot gas, either hot or diluted with varying amounts of primary air. Hot secondary air is propelled through the cooling zone, providing more oxygen for combustion and the necessary draft. The thermal efficiency of the kiln is low and heat consumptions about 6688kJ/kg[6]. Also the producer gas kiln spawns more complex environmental problems including disposal and treatment of a sulphurous liquid discharge than possible with any other type of kiln.

2.1.5 Gas Fired centre-burner Kiln

This is an advanced shaft kiln. The fuel may be natural gas or atomized fuel oil. The introduction of centre burners on two or four sides of the kiln into the calcining zone improved kiln performance, fuel efficiency and lime quality. Centre burners provide a more even temperature to all sections of the calcining zone. This feature reduces fuel consumption and the capacity of the kiln greatly increased from 2.5 to 5 tone to 10 – 20 tons/da/m^2 [7], put differently the same size of cylindrical shell that produced 15 tons/day in standard shaft kiln is able to produce 40-60 tons/day.

2.1.6 Large Capacity Mixed Feed Kiln

Inspite of producing an inferior quality of lime on an average, mixed feed kilns have developed with new refinements and improvements [8]. This is employed when the quality of lime is relatively unimportant. In order to achieve maximum thermal efficiency, a dense, metallurgical grade of coke or low reactivity or anthracite coal both of which are low in ash and volatiles are admixed intimately with the kiln feed at a predetermined proportion. This method is far superior to traditional mixed-feed charging of alternate beds of fuel and stone since it obviates excessive localized temperatures with resultant over burning and promotes more even heat distribution throughout the whole kiln cross section. About 85% of theoretical thermal efficiency based on how heat value and a capacity of 270 tons/day have been reported [9] for this type.

2.1.7 Parallel-Flow Kiln

The parallel-flow regenerative vertical kiln of proved capacities ranging from 100 to 600 metric tons/day on an average, appears to possess the best and most consistent thermal efficiency of any kiln in history[10]. It operates with either two or three independent shafts within one large vertical, refractory0lined shell and utilizes alternative parallel-counter flow through the stone charge in the other shafts. Thus the upper portion of each shaft become the heat generator. Calcinations occurs at present intervals of 10-20 mins. Quicklime is 66 – 93^{0}c. Fuel usually natural gas in introduced from above downward through multiple firing lances in parallel flow. As evidence of extremely high degree of heat recuperation exist gases reportedly have temperatures of about 12^{0}c [11].

2.1.8 Double-Inclined Kiln

In this type the stone moves downward gravity, however the path is tortuous. The dual inclination turns the stone over for the more uniform calcinations. Opposite each inclined wall are three short cylindrical combustion chambers and above each chamber is an arched roof that helps to retain and concentrate the temperature in the dual calcining zones. Fuel consumption averages 4389-1681kJ/kg. The kiln can be fired with combined fuel mixtures and this optimizes thermal efficiency than a single fuel type.

2.1.9 Annular (Ring) Kiln

The internal kiln construction features an upper and lower inner cylinder that functions in handling the process gases. Thus an annular opening or ring inside the kiln's periphery is formed to receive the limestone charger. The rate of stone lime movement is controlled by positive displacement type dischargers. The cooling lime is withdrawn at programmed intervals by a vibrating feeder. Total lime residence time in the kiln is 25-33 hrs. The kiln can be fired either by a natural gas or oil and operates under negative pressure with combustion air derived from three sources: primary air ascending from the cooling system, secondary inflows as primary air for the burners and compressed air for the injectors. The hot injector air received from the recuperate at 454^{0}C provides velocity for recirculation of the process gases[12].

3.0 Materials and Methods

The models used to describe the transport phenomena and the simplifying assumptions usually employed, are as follows.

3.1 Reaction Model

The calcination is modeled based on the reaction limiting step. Various models have been proposed by many workers [13]. These models are based on the following resistances to calcination.

a) Chemical reaction at the interface between the unreacted core $CaCO_3$ and the product CaO.
b) Diffusion of $C0_2$ through the calcine (CaO) away from the reaction zone.
c) Conduction of heat to the reaction interface.
d) Diffusion of $C0_2$ away from the particles; into the particulate phase**.**

Baker (1962) [14] has investigated the equilibrium reaction of limestone dissociation.

$$CaCO_3(s) \longrightarrow CaO(s) + CO_2(g) \qquad (1)$$

and found that its equilibrium constant K~ (in units of atmospheres) is best given by ,

$$\log_1 Kp = \log Peq - 8303/T + 7.079 \qquad (2)$$

Simple conservation consideration (Hills, 1968) show that the rate of decomposition of the carbonate sphere is

$$n=4\pi r^2 \rho_{co2}\, dr/dt \qquad (3)$$

The calcining particle is treated as a sphere if dr/dt is constant the identity in Equation (2) shows that reaction rate per unit area of the reaction interface is also constant. The approximate constancy of dr/dt has led many workers to propose that the reaction is controlled by a chemical step at the reaction interface.

lngreham and Marrier (1963) [15] suggested that the step limiting the reaction is the diffusion of CO_2 through a thin layer of active lime. Hayatte, Cutter and Wardsworth (1958) [16] proposed a mechanism similar to the surface catalysis of gas reactions involving the reaction of carbonate, active lime and absorbed carbon dioxide on the interface. Satterfield and Feakes (1959) [17] have also proposed a chemical control mechanism which involves the cracking of the original carbonate lattice and concluded that heat transfer is critical to the rate of reaction especially at high temperatures. Haslam and Smith (1928) [18] and Narsimhan (1961) [19] have also advanced theoretical treatments for the reaction which assume it to be exclusively heat transfer controlled. Hills (1968) [3] also investigated the reaction mechanism and put forward some models. These models indicate that the reaction is limited by heat and mass transfer.

The reactor model of an operating kiln is dependent on the nature of operation. Kilns are modelled essentially as moving bed reactors or fluidized bed reactors. The overall model for the kiln is composed of, in principle layer models as described above which are coupled via the material and energy balances of the relevant gas flow (fuel, air and exit gas), and solid flow (limestone and burnt lime). Each layer of limestone in the feed batch is during its passage through the kiln exposed to continually varying ambient conditions which determine its temperature and thus the burning process. If the behaviour of these ambient conditions for the stabilized operating state of the kiln can be ascertained, then the temperature and burning behaviour of the lime will be determined.

3.2 Transport Phenomena

The flow of materials around a lime kiln is shown in a block diagram as in Figure 1

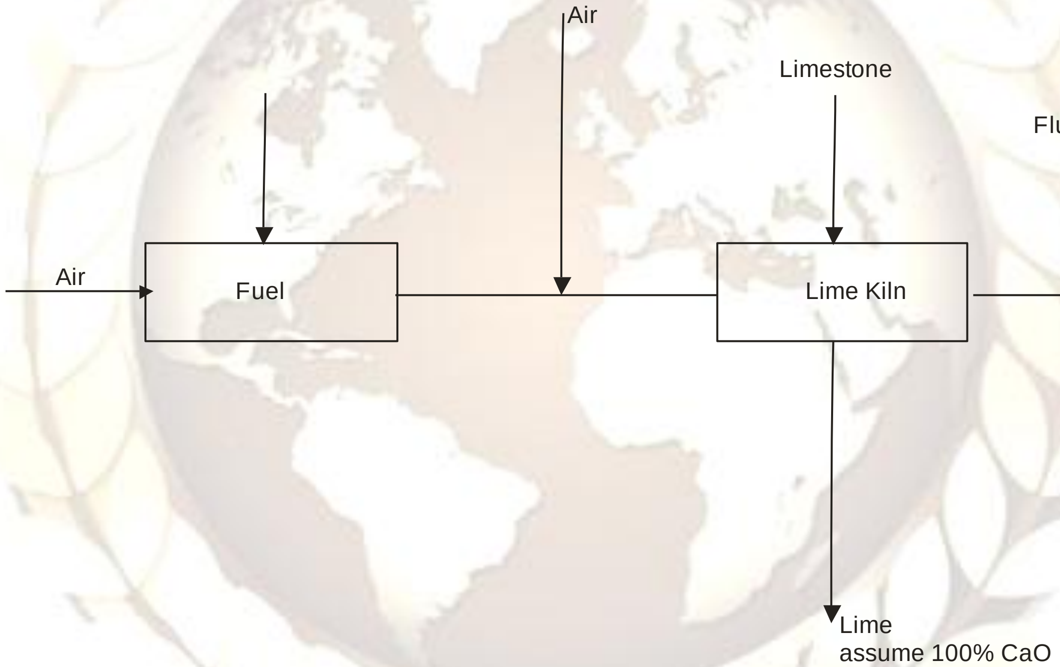


FIGURE 1 MATERIALS FLOW AROUND A KILN

The scheme shows that a mixture of air and fuel are burnt and the hot flue gases from the burners enter the kiln which is charged with limestone. The limestone is decomposed into lime and carbon dioxide. The liberated carbon dioxide and the combustion gases leave as kiln flue gases and the lime leave as the product. Heat content of the combustion gases are abstracted by the limestone in the burning zone of the kiln to effect the calcination of the limestone. The remaining heat content of the combustion gases that are not abstracted in the burning zone are also abstracted by limestone in the preheating zone to effect the preheating of the limestone charge before it enters the burning zone of the kiln. Cool air is also passed through the cooling zone to abstract residual heat from hot burnt lime and this contributes to the preheating of either the limestone in the preheating zone or is employed in preheating of fuel when heavy fuels are employed in the process.

The process thus entail:

a) Combustion of fuel .
b) Exchange of heat between limestone charge and hot combustion gases.
c) Chemical decomposition of limestone into lime and carbon dioxide.
d) Exchange of heat between hot burnt lime and cooling air.
e) Loss of heat from flue gases.
f) Loss of heat from the kiln shell.
g) Flow of limestone in the kiln .
h) Flow of the gases in the kiln .

The heat transport follow the three basic means of heat transmission namely;

a) Conduction
b) Convection
c) Radiation

The layer models employed to describe the prevailing transport phenomena for the kiln are as follows[20]:

3.2.1Conduction Heat Transfer

Fourier’s law is the fundamental differential equation for heat transfer by conduction;

$$dq/d\theta = -kA(dt/dx) \qquad (4)$$

Where

$dq/d\theta$ = rate of flow of heat
A = area at right angles to the direction in which heat flows
$-dt/dx$ = rate of change of temperature with distance
k = conductivity constant

For conduction through multi layer transport of heat , the heat transfer is given by

$$Q = \Sigma \Delta t / R_T = (t_1 - t_2) / R_T \qquad (5)$$

Where R_T is the overall resistance and is the sum of the individual resistances in series as follows;

$$R_T = R1 + R2 + \ldots\ldots\ldots\ldots + Rn \qquad (6)$$

When a wall is constructed of several layers of solids, the joints of adjacent layers may not perfectly exclude air spaces and these additional resistances should not be overlooked.

3.2.2 Heat Transfer by Convection

The temperature gradients in both the fluid and the adjacent solid at the fluid- solid interface may also be related to the heat transfer coefficient as

$$dq = hi\, dAi\, (t_1 - t_3) \qquad (7)$$

If q is a linear function of Δt and h is constant then

$$q = \frac{hA(\Delta t_{in} - \Delta t_{out})}{\ln(\Delta t_{in} / \Delta t_{out})} \qquad (8)$$

where Δt factor is the logarithmic mean temperature difference between the wall and the fluid

The heat capacity of the gases is described by the equation;

$$\text{Heat capacity} = C1 + C2T + C3T^2 + C4T^3 + C5T^4 \qquad (9)$$

Where C1, C2,C3,C4 and C5 are polynomial constants of the gas under consideration.

3.2.3 Radiative Exchange between surface of solids

According to Kirchoffi's law, the emissivity and absorptivity of a surface in surroundings at its own temperature are same for both monochromatic and total radiation. When the temperatures of the surface and its surroundings differ the total emissivity and absorptivity of the surface often are found to be different but because absorptivity is substantially independent of irradiation density, the monochromatic emissivity and absorptivity of surface are for all practicable purposes same.

Consider radiative exchange between a body of area A_1 and temperature T_1 and black surroundings at T_2. The net interchange is given by

$$Q_{1=2} = A_1 (\epsilon_1 \sigma T^4 - \alpha_{12} \sigma T_2^1) \qquad (10)$$

The value of ϵ_1 (α_{12} the absorptivity of surface A_1 for blackbody radiation of T_2) is the Area under the curve of ϵ_λ versus f the latter read as a function of λT_1 (or λT_2)

The radiation from a flame is due to radiation from burning soot particles of microscopic and Submicroscopic dimensions from suspended larger particles of coal, coke or ash from the water vapour and carbon dioxide in hot gaseous combustion products. The contribution of radiation emitted by combustion process itself, so called chemical luminescence is relatively negligible.

3.2.4 Mass Transfer

During the process of calcination of limeastone , the reaction progresses via shrinking core kinetics. As the reaction progresses the carbon dioxide liberated is transported from the core through the calcine to the outer surface of the reacting particle and then to the bulk gas phase.

Simplified flux equations of Fick's law can be used for unidimensional steady state problems with binary mixtures. The boundary conditions represent the compositions represent the compositions X_{AL} and X_{AR} at the left hand and right hand sides of a hypothetical layer having thickness Δz. The principal restriction of the following equations is that concentration and diffusivity are assumed to be constant.

For equimolar counter diffusion $N_A = -N_B$

$$N_A = mJ_A = - D_{AB}\, c\, dxA/dz = D_{AB} c(X_{AL} - X_{AR}) \qquad (11)$$

For unimolar diffusion ($N_B = 0$)

$$N_A = mJA + X_A N_A = \frac{D_{AB}}{\Delta z} c \ln \frac{1- X_{AR}}{1 - X_{AL}} \qquad (12)$$

3.2.5 Flow through Beds of Solids

The hot combustion gases flow into the kiln through the combustion chambers through the bur

For isothermal compressible flow of a gas with constant compressibility factor z through a packed bed of granular solids is given by;

$$P_1^2 - P_2^2 = \frac{2ZRG^2T}{M_w}[\ln v_2/v_1 + 2fmL(1 -6)^{3-n}/ \varphi^{3-n} \epsilon^3 Dp] \qquad (13)$$

where

P_1 = upstream absolute pressure
P_2 = downstream absolute pressure
R = gas constant J/kgmol.K
M_W = molecular weight
V_1 = upstream specigic volume of gas

V_2 = downstream specific volume of gas
G = mass flux $kg/m^2.s$
T = absolute temperature K
L = length m
Dp = diameter of particle m
f = fanning friction factor
φ = energy dissipation ratio per unit volume $J/m^2.s$
ϵ = void fraction

3.2.6 Combustion

The amount of oxidant (oxygen or air) just sufficient to burn , the carbon , hydrogen and sulphur in a fuel to carbon dioxide is the theoretical or stoichiometric oxygen or air requirement.
The chemical equation for complete combustion of a fuel is

$$C_XH_yO_zS_w + (4x + y -2x + 4w/4)\ O_2 = x\ CO_2 + (y/2)H_2O + WsO_2 \qquad (14)$$

X, y , z and w being the number of atoms of carbon, hydrogen, oxygen and sulphur respectively in the fuel.

The volume of theoretical oxygen(at 0.101MPa and 298K) needed to burn be calculated from the ultimate analysis of the fuel is as follows;

$$24.45(C/12 + h/4 – O/32 + S/32) = m^3\ O_2/\ kg\ fuel \qquad (15)$$

Where C,H,O and S are the decimal weights of these elements in 1kg of fuel . The mass of oxygen (in kg) required can be obtained by multiplying the volume by 1.31. The volume of theoretical air can be obtained by using a coefficient of 116.4 in the above equation in place of 24.45.

3.2.7 Assumptions

The following simplifying assumptions can be employed in the development of kiln simulation model as in Table 1;

TABLE 1 MODELS USED IN KILN SIMULATION PROCESS MODEL

Phenomenon	**Prevailing conditions**
Heating of gas	Polynomial expressions for thermal capacity.
Data for gas	Ideal gas
Pressure Loss	Fanning equation and Poiseuilles law
Heat Transfer Lime-lime	Uniform temperature level in Kiln charge, no direct lime-lime transfer from layer to layer is assumed.
Heat Transfer gas-wall	Heat transfer coefficient calculated from equation based on the prevailing conditions for gas-lime interface. The radiations effect is neglected.
Heat transfer lime-wall	Head transfer coefficient of the lime surface is employed en the heat transfer equation.
Heat loss through kiln wall	Heat transfer equation for composite wall. The external surface conductivity is assumed constant.
Material transfer Gas-lime	This is usually neglected
Material transfer lime - gas	This is calculated as plain mixing (CO_2 mixed with the gas)

3.3 Experimental

A laboratory muffle furnace of dimension 30cm by 20cm manufactured by Philip Harris UK was used. The furnace is electrically heated with temperature regulator thermostatically controlled. The temperatures used in the study are usually set and sized limestones are burned in very clean nickel crucibles. The weight of the samples are monitored over the period of calcination.

The weights were measured with electronic balance to 0.01. This was by withdrawing the sample from the furnace at intervals and taking the weight. The measurement was done very fast. This was to ensure that the sample readily assume the temperature of the furnace. The opening of the furnace door at intervals ensures the sweeping of air through the furnace to keep the composition of the gas phase around the sample constant during each sun. The sensitivity of the experimental result was improved using the “First degree least squares with “five points smothering technique”. This approximates the readings of the sample weight taken to a continuous reading at discrete intervals as would have been necessary. The rate of reaction was obtained by numerical differentiation of the sample weight using “Lagrange’s five point interpolation formula [21], Hovanessian (1976)) [22]. The results obtained were used to obtain the calcination constants. Then the results were extended to a range of pressure and particle size in order to obtain a simulation result.

4.0 Discussion

The results obtained from the simulation of the calcination of the limestones are as follows;

4.1 Variation in Particle Size

From Figures 2- 4, at fixed temperatures the time for total calcination of a particle increases as the particle size increases. The time for total calcination also increases with increase in pressure. Since the reaction approximates shrinking core kinetics the reaction readily progress inwardly from the particle surface.

This progression is time dependent thus with increase in particle size the time for complete calcination increases. The Jakura limestone has higher calcination time than Ukpilla limestone, at specific temperature and pressure. This implies that Ukpilla limestone will have higher kiln through put during burning.

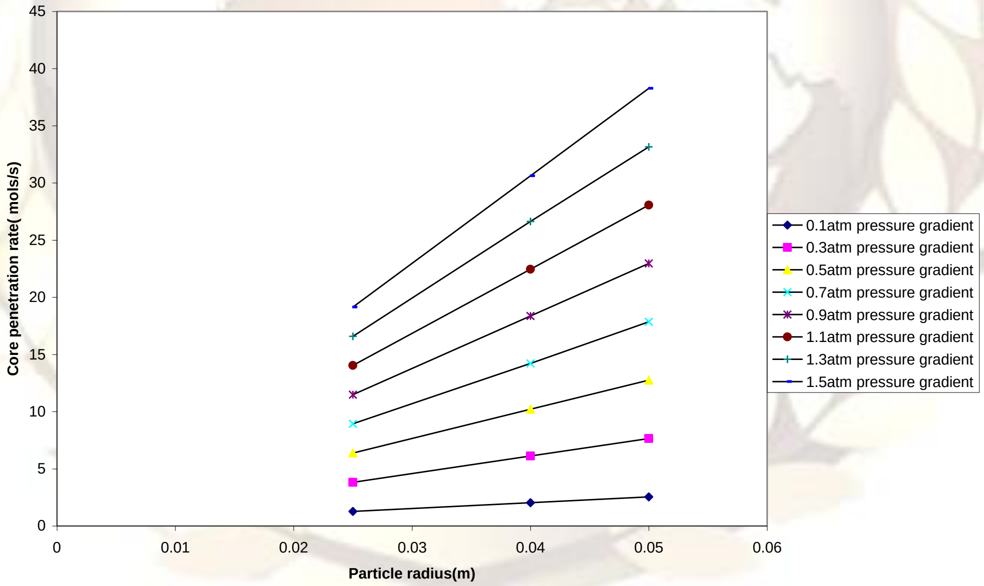


Figure 2 Variation of Core penetration rate with Particle radius

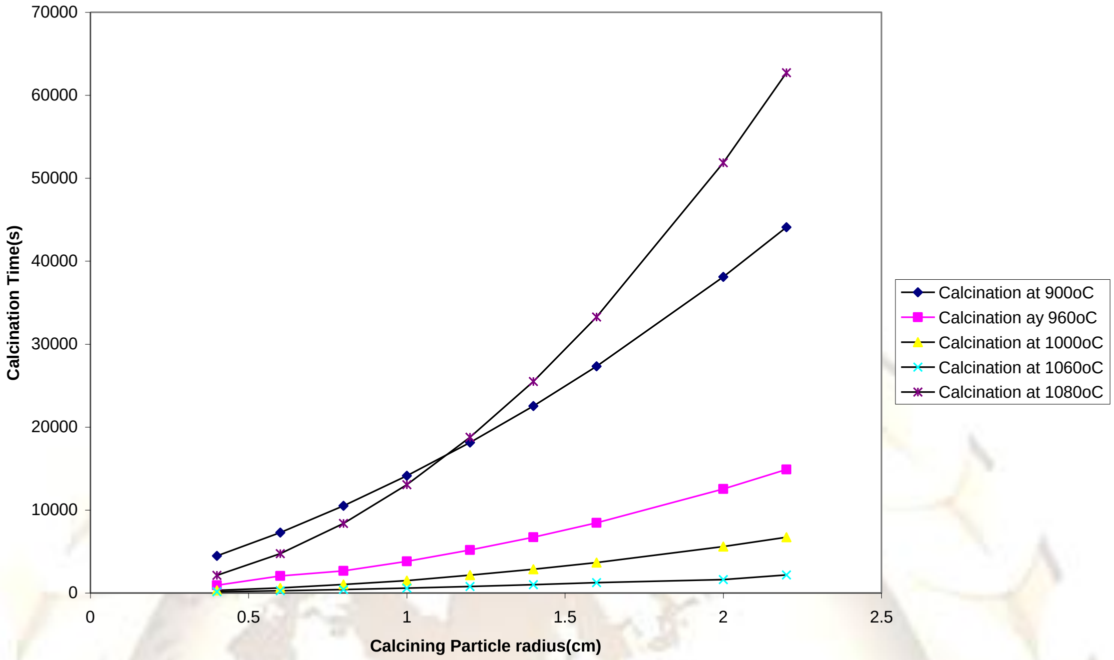
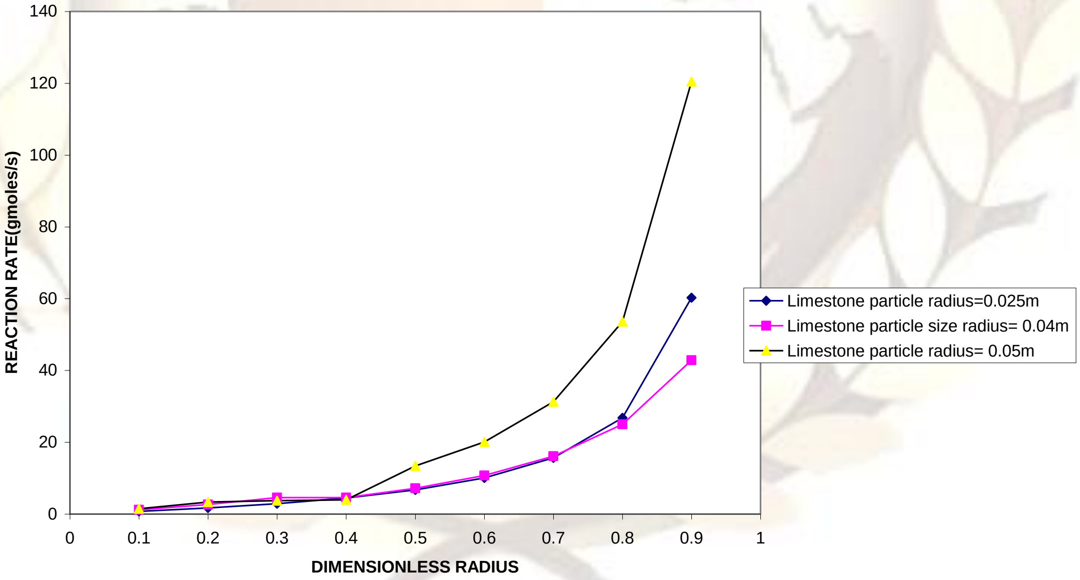


Figure 3 Variation of Calcination Time with Particle radius(Jakura Limestone)



FGURE 4 VARIATION OF REACTION RATE WITH DIMENSIONLESS RADIUS(HEAT AND MASS TRANSFER LIMITED REACTION)

4.2 Variation in Temperature

The time for total calcination decreases with increase in temperature up to 1060^{0}C for both limestones. The calcination reaction an endothermic reaction thus higher temperatures which implies higher energy content of the surroundings will necessarily increase the rate of the reaction. The shrinking core kinetics mode suggests that the transport of mass and heat to the unburned core is through the lime layer. At higher temperatures the lime layer is over burned thus decreasing; the transport of CO_2 from the reaction interface.

The decrease in transport of CO_2 leads to higher pressure of CO_2 at the reaction interface which Increases the backward reaction which is carbonation. The Ukpilla limestone has generally a lower calcination time than Jakura limestone at the same temperature levels. This is shown in Figures 5 and 6

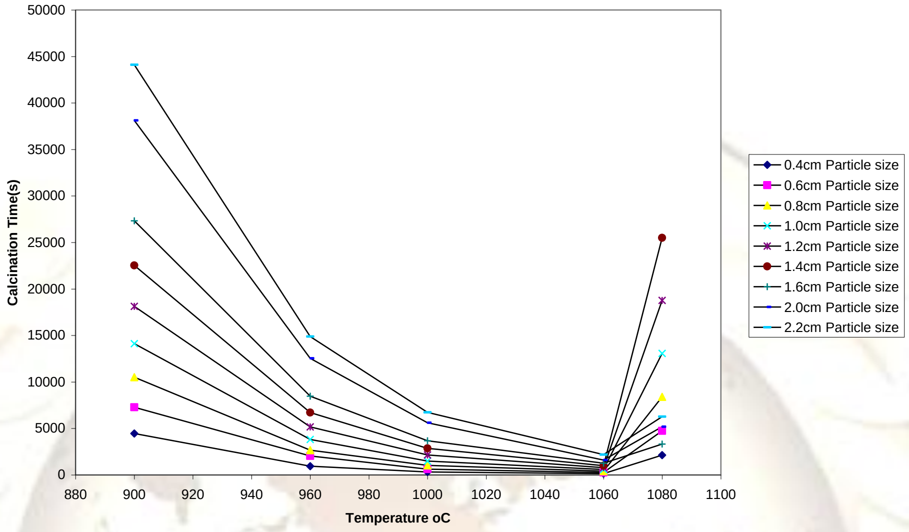


Figure 5 Variation of Calcination Time with Temperature for Jakura Limestone

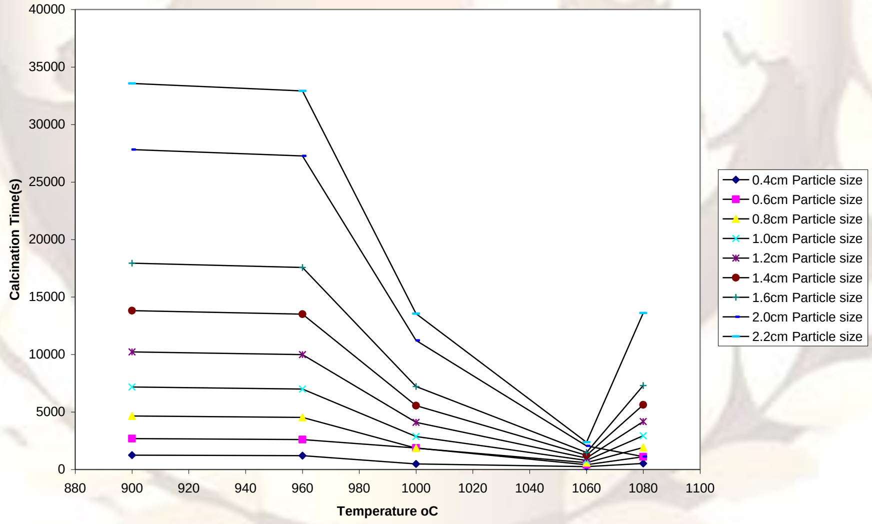


Figure 6 Variation of Calcination Time withTemperatuire for Ukpilla Limestone

4.3 Variation in Pressure

The time for calcination increases with increase in pressure. The explanation of this trend is that limestone calcination is an equilibrium reaction. High pressure in the calcination environment implies high CO_2 partial pressure. This condition increases carbonation reaction is the reverse of the decomposition reaction. This is shown in Figures 7 – 9.

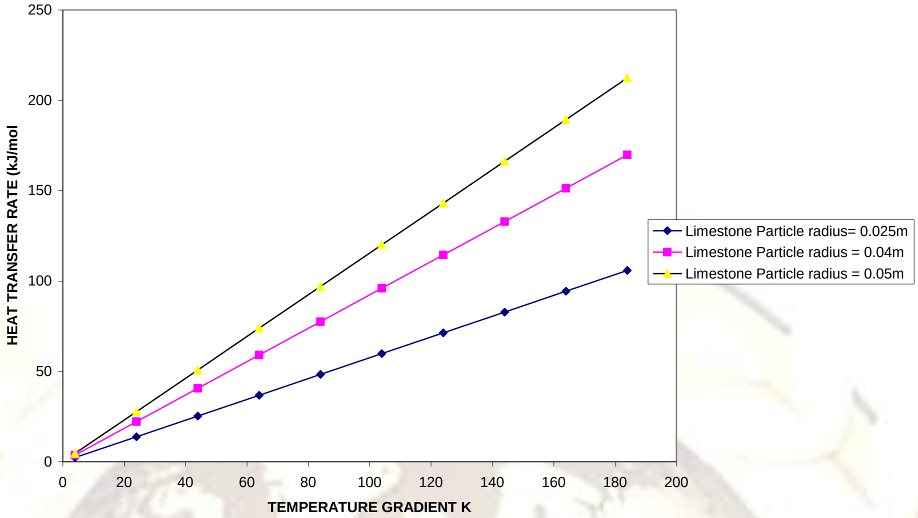


FIGURE 7 VARIATION OF HEAT TRANSFER RATE WITH TEMPERATURE GRADIENT ACROSS THE LIME LAYER(HEAT AND MASS TRANSFER LIMITED)

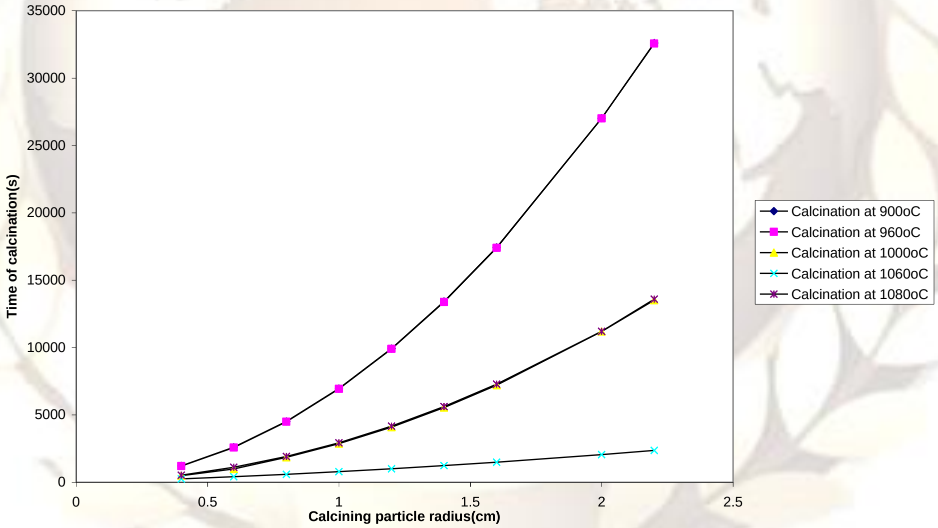


Figure 8 Variation of calcination time with particle radius at -345mmH20(Ukpilla Limestone)

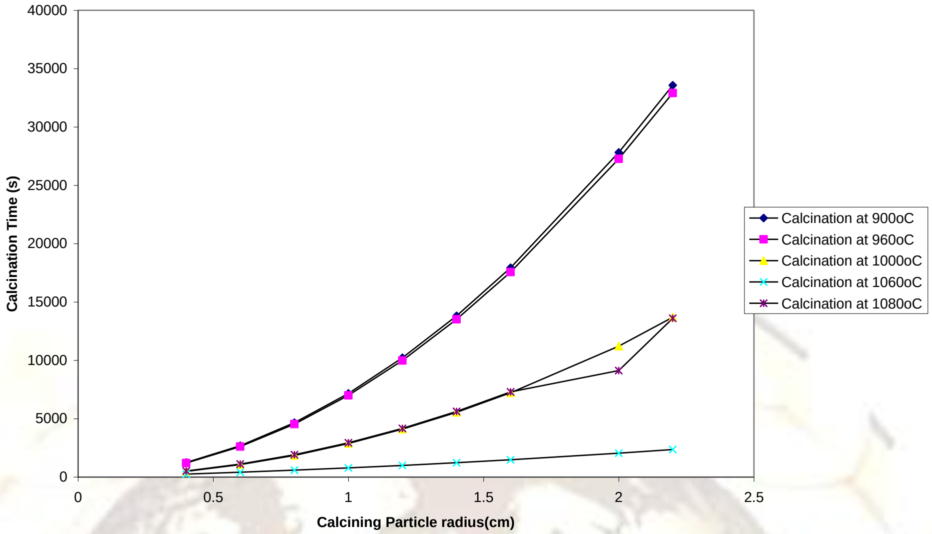


Figure 9 Variation of Calcination Time with Particle radius at -100mmH20 (Ukpilla Limestone)

4.4 Intra Particle kinetics

From Figure 10 , the time for complete calcinations of particles is more than that of smaller sizes. The rate of core penetration that will achieve small times of complete calcinations is greater for bigger limestone sizes hence to burn large particles more stringent conditions of temperature will be required. However this will have a negative impact on the quality of the lime because high calcinations temperatures will lead to hard burning of the lime. Hard burned limes are usually less reactive because of the occulusion of the pores leading to loss reaction surface.

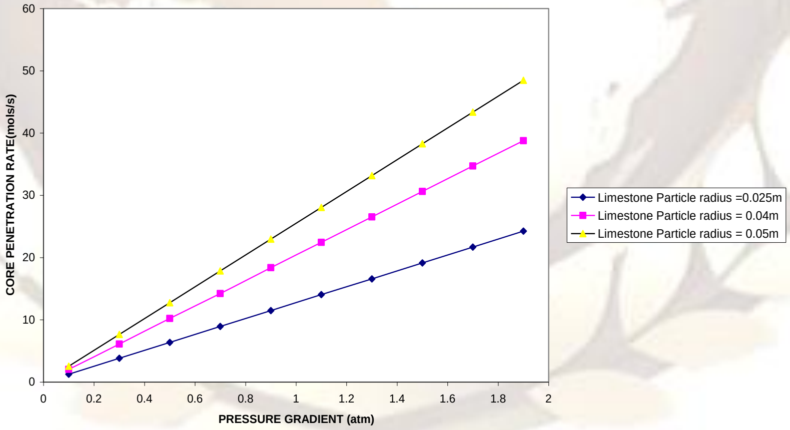


FIGURE 10 VARIATION OF CORE PENETRATION RATE WITH PRESSURE GRADIENT ACROSS PRODUCT LAYER (HEAT AND MASS TRANSFER LIMITED REACTION)

Conclusion

This work has shown the various phenomena that prevail in the kiln during operation and these determine the design and operational demands of efficient lime production process. The development of a simulation model for lime production is dependent on the deployment of the layer models as outlined in the work with the simplifying assumptions. This work will further the development of efficient lime production processes that will meet the increasing need for high quality products and the increasing needs of large industrial consumption. This will also aid in optimizing the energy demands of the processes in the face of global energy crisis.

From the simulation of the calcination model for Jakura and Ukpilla limestones, it can be deduced that:

(1) Calcination time decreases with increase in particle size
(2) Calcination time decrease with the increase in temperature up to 1060^{o}C but Increases with increase in temperature above 1080^{0}C.
(3) Calcination time increases with increase in pressure.

These trends are essential for the economic operation of a kiln designed for the calcination of these llmestones.

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