[ED05] Firing of Limestone in JPN Pilot Plant

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Introduction

Limestone is a natural occurring mineral throughout the world. It is widely used in the cement manufacturing, construction and building, metal refining processes, quicklime industries, agricultural industries, flue gases treatments, etc. The chemical composition of the limestone varies from one region to the other region, but it mainly consists of calcium carbonate. In the Peninsular Malaysia, 7 billion tonnes reserve of limestone has been identified by the Geological Survey Department, Malaysia. It is found that 55% of the limestone in Peninsular Malaysia contains 95% calcium carbonate (Sulaiman, 1996). It is feasible to be used as raw materials to produce quicklime.

The dissociation of limestone is an energy intensive process. During dissociation process, the carbon dioxide gas is expelled from the limestone to produce quicklime at the theoretical dissociation temperature of 900°C.

$$CaCO_{3\,(s)} + Heat \rightarrow CaO_{(s)} + CO_{2\,(g)} \uparrow$$

Few factors affecting the rate of dissociation have been outlined in the literature. These factors are source of the limestone (i.e. structure, composition, and density), particle size distribution, shape of particles, firing temperature, rate of heating, and partial pressure of carbon dioxide gas in the kiln (Boynton, 1980; Wingate, 1985; Oates, 1998). Recently, Moropoulou *et al.* (2000) studied the reactivity of the quicklime produced from various source of limestone. He reported that the quicklime produced from inhomogeneous grain size limestone with less compact structure was more reactive than the homogeneous fine grain size limestone with more compact and harder structure. He also found that quicklime produced at 900°C showed the higher reactivity and surface area compared to the quicklime produced at other higher temperatures.

However, no studies have been reported to the temperature profile of the packed bed in the kiln, which directly affects the rate of dissociation and the quality of the produced quicklime. Therefore, in this work, an attempt was made to design and fabricate JPN Pilot Plant and to study the various parameters for firing of limestone.

Material and methods

The high calcium limestone was selected from Kinta Valley quarry in Perak. The large limestone particles were crushed into small pieces. Each particle size was 33 mm of sphericity 0.64.

The limestone was fired in the pilot plant in Figure 1. This pilot plant consisted of firing kiln fitted with liquefied petroleum gas (LPG) burning system, spiral jacketed baffle cooler, and air intake system. The combustion chamber in Figure 2 was designed specially according to the flame propagation in order to provide uniform heat distribution to the whole packed bed. One thermocouple was fitted in the combustion chamber and one in the outlet of the firing kiln to measure flame temperature and the outlet flue gases temperature respectively. The firing chamber of 25 cm diameter, fitted with 16 thermocouples, located across and along the chamber at equal distances of 5 cm to record the temperatures in the chamber.

One to six layers of limestone, weighed 1.49 kg to 8.94 kg, were packed in the firing chamber. Each layer was made up of 58 limestone particles, as shown in Figure 3.

Air/fuel ratio was 32.4/1 in order to generate 20 kW heat for the packed bed. The temperatures were recorded each 5 minutes over various firing time of 1, 1.5, 2, 2.5, and 3 hours. The conversion of limestone to quicklime and carbon dioxide was calculated.

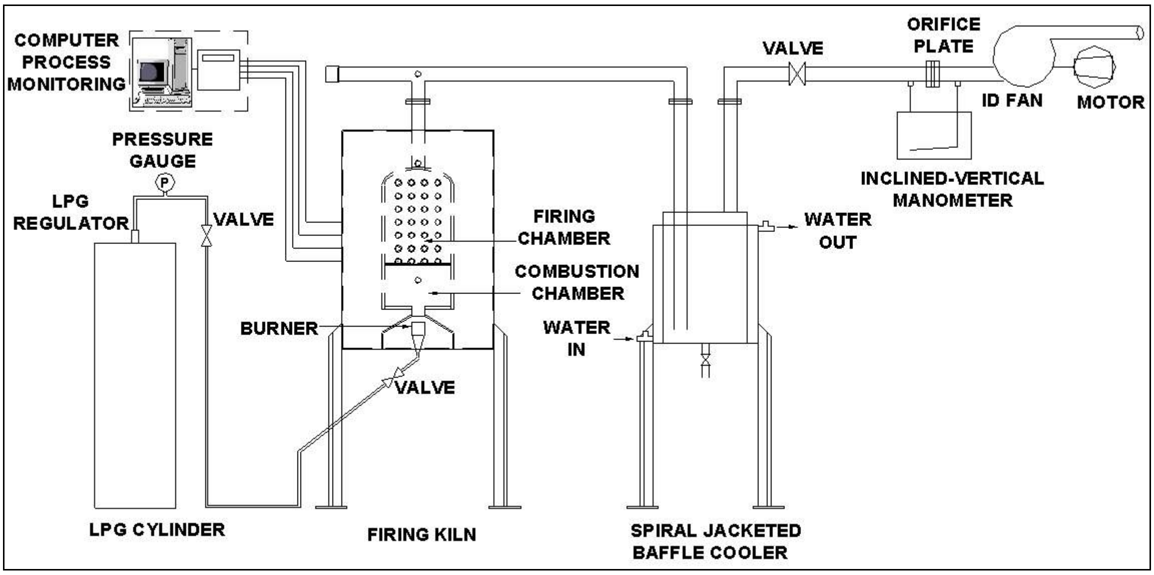

FIGURE 1 Schematic diagram of the JPN Pilot Plant for firing of limestone to produce quicklime.

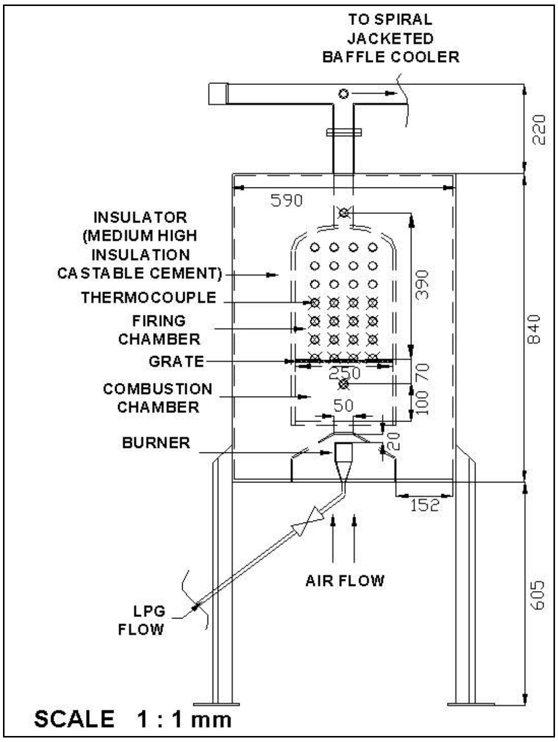

FIGURE 2 Schematic diagram of the firing kiln.

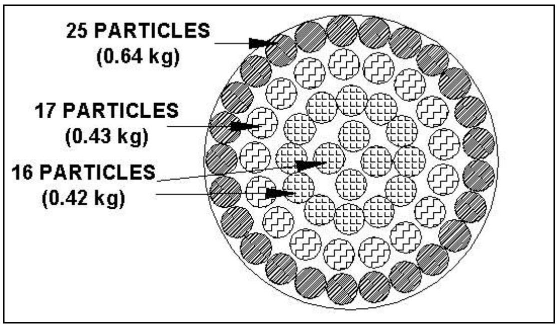

FIGURE 3 Arrangement of the limestone particles for each layer.

Results and discussion

Figure 4 shows the average temperatures for the flame, packed bed and outlet flue gases versus firing time over a period of 3 hours. The temperature variation of flame showed to be higher than the packed bed while the outlet flue gases temperature showed to be the lowest as expected. The effective temperature of the flame to achieve 900°C for the packed bed can be deduced from the graph to be 1130°C after the elapse of almost 1 hour of time from the start up of the firing. In the following firing time of 2 hours, the flame temperature increased to 1280°C. A conversion of 87% for the limestone from the packed bed curves can be also deduced to be at temperature of 1130°C after the elapse of 3 hours of firing time. The corresponding temperatures of the outlet flue gases from the kiln deduced from the graph were varied from 450°C to 690°C over this period of 1-3 hours. This indicated appreciable sensible heat was leaving the kiln. That could be used as the waste heat for other purposes.

Figure 5 shows the average temperatures for bottom layer, middle layer, and top layer of the packed bed versus firing time for firing 5 layers of limestone. The average temperatures of each layer decreased from the bottom layer to the top layer in the packed bed. The average temperature difference between the bottom layer and the top layer of the packed bed was almost 400°C over 0.5-3 hours of firing time. At the half an hour of firing time, the average temperatures of the bottom layer, middle layer, and top layer of the packed bed increased rapidly from ambient temperature to 930°C, 790°C, and 530°C respectively. It showed that only the average temperature of the bottom layer of the packed bed was higher than the theoretical limestone dissociation temperature of 900°C. Therefore, the significant dissociation of limestone was only in the first or first two layers of limestone. The higher layers of the limestone were not dissociated or the dissociation might have taken place on the surface of the limestone because of the temperature below 900°C. After 1.1 hours of firing time, the average temperature of the middle layer of packed bed had approached to 900°C with the conversion of 24%, but it had increased rapidly to 84% in the second hour of firing time as shown in Figure 7. When the packed bed was fired for 3 hours, the average temperatures of the bottom layer, middle layer and top layer of the packed bed reached to 1250°, 1200°C and 880°C respectively. However, average temperature of the top layer of the packed bed still was under 900°C. It caused that the conversion of the top layer to be only 50%, compared to the other 4 layers of limestone with the conversion of more than 90%. It showed that the firing of limestone at the temperature below 900°C was not feasible to produce quicklime for commercial purposes.

Figure 6 shows the average temperature profiles over various periods of firing in packed bed versus radial position across the firing chamber for firing 5 layers of limestone. The curves were concave downwards. This indicated that the conversion of the limestone to the quicklime and carbon dioxide was endothermic reaction. The concavity of the curves was reduced from half an hour to 3 hours of firing time. The curve after 3 hours of firing become almost straight line, indicating the steady state condition for the endothermic reaction with the uniform average temperature. The average temperature across the radial of the whole packed bed over 3 hours of firing time was 910°C with the conversion of 87%.

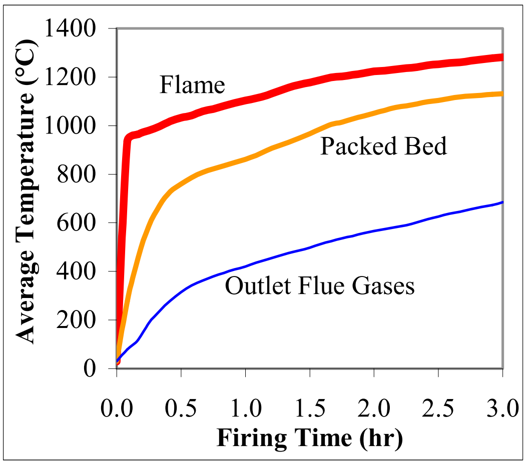

FIGURE 4 The average temperatures for the flame, packed bed and outlet flue gases versus firing time for firing 5 layers of limestone.

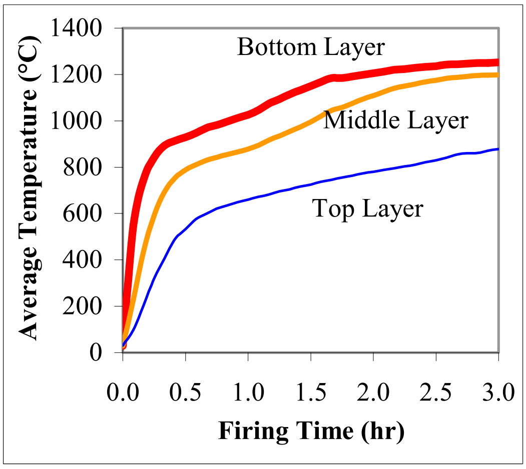

FIGURE 5 The average temperatures for bottom layer, middle layer, and top layer of the packed bed versus firing time for firing 5 layers of limestone.

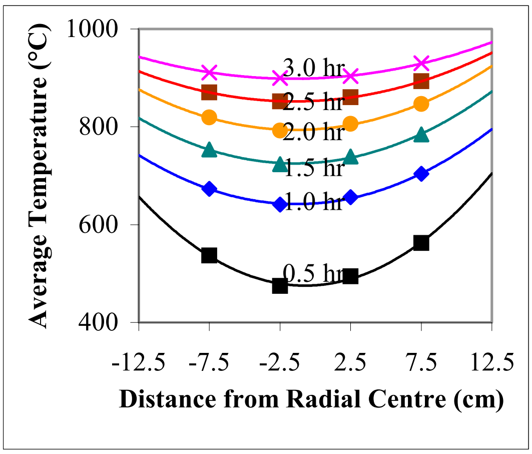

FIGURE 6 The average temperature profiles over various periods of firing in the packed bed versus radial position across the firing chamber for firing 5 layers of limestone.

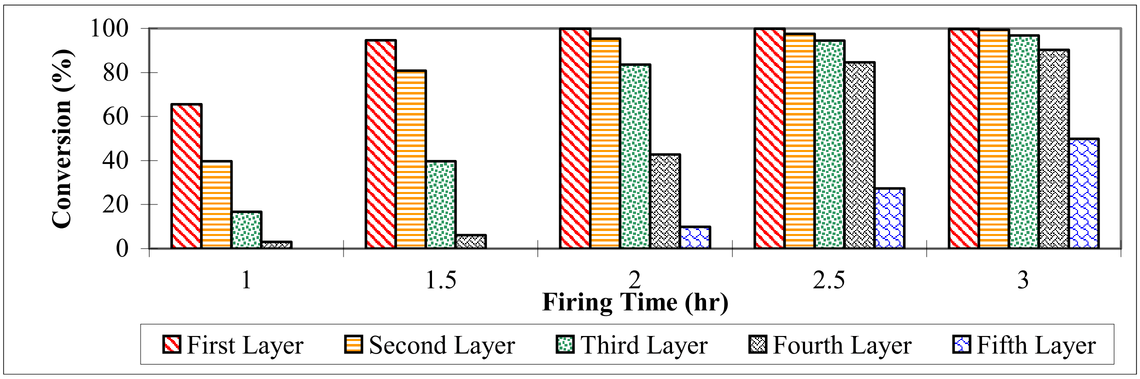

FIGURE 7 Conversion of limestone for each layer when 5 layers of limestone were fired at various firing time.

Figure 8 shows that the conversion of limestone versus number of layers of limestone particles after 3 hours of firing time. The maximum conversion of 95% was deduced to be at 4 layers of limestone particles. This could be explained by that 20 kW of dissipated heat by the burner to the various numbers of layers with different heat transfer controlling mode. Meaning that the one layer of limestone was heated by the radiation heat transfer mode more than convection and conduction. This was mainly due to the high voidage of 61%, which allowed the hot gases to pass through in a very short time, causing insufficient time for convection. As the number of layer increased, the corresponding number and weight of the particles increased, causing the packed bed to be denser and denser to a voidage of 44% at 4 layers of limestone particles. Therefore, the convection and conduction heat transfer modes were playing a great role for heating up the packed bed faster to above 900°C, while the temperature rise of the one layer had gone up very slowly to 900°C, confirming the above discussion.

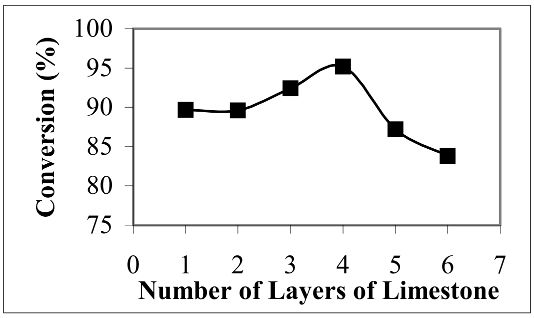

FIGURE 8 The conversion of limestone versus number of layers of limestone particles after 3 hours of firing time.

Conclusions

Maximum conversion of 95% was obtained by 20 kW of heat over 3 hours in JPN Pilot Plant for firing of limestone particles of 4 layers, weighed 5.96 kg, average particle size 33 mm, and sphericity 0.64.

Acknowledgements

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