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FUTURE CEMENT
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Pilot scale manufacture of limestone calcined clay cement : The Indian experience

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Pilot production of a new type of ternary blend cement, containing 50% clinker, 30% calcined clay, 15% crushed limestone and 5% gypsum (LC3), was carried out in India. The raw materials required to produce the cement were found to be easily available in the quality required for the production. Calcination of clays was carried out in static kilns used to fire potteries and ceramics. Grinding and blending of the cements was carried out at a cement grinding unit. The blends produced were tested in the laboratory and building materials were produced using the cement. Good results were obtained from the blends despite the sub-optimal conditions of production of the cement, demonstrating the viability and robustness of the technology.

Keywords: *Limestone; calcined clay; ternary cement; low clinker; low carbon.*

1. INTRODUCTION

Although cement is known to be one of the largest sources of anthropogenic CO_2 emissions around the world, due to the composition of the earths crust and the limited availability of various raw materials, the options to reduce the impact of construction on the environment are limited [1,2]. It has recently been shown that the substitution of clinker by a mixture of calcined clay and limestone can give better strengths than OPC even at a low clinker factor of 0.5 [3]. Calcined clays, especially of the kaolinitic type, have been known to be a pozzolanic material for long and several standards around the world, e.g. Indian and Brazilian, have permitted their blending in cements and concretes [4,5]. Uncalcined limestone is also commonly used to replace up to 35% clinker in cements, with research showing that replacement of up to 10% of clinker with crushed limestone has little negative effect on the properties of concrete [6].

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The material presented in this article attempts to combine the advantages of using calcined clays and limestone in cements to achieve clinker factors as low as 40% in practice. The initial laboratory studies of the material in Switzerland and Cuba have demonstrated the synergetic effect of clinker, limestone and calcined clays, leading to the formation of carbo-aluminates that continue to reduce the porosity and therefore increase the strength of cement pastes, even as the pozzolanic reaction between the hydration products of clinker and calcined clay continues [3]. This blend of cement has been named LC3 for Limestone Calcined Clay cement. More details on the composition and chemistry of these cement blends can be found elsewhere [3,7]. The feasibility of the production of LC3 blends at an industrial scale using easily available technology is demonstrated in this article.

This article discusses the identification of raw materials for production of this ternary blended cement at a pilot scale in India, the processing of these materials and the final production and testing of the cement produced. In order to demonstrate the ease of production of the material and the robustness of the technology, the lowest possible level of sophistication was used for this process of production.

2. RAW MATERIAL IDENTIFICATION

Raw materials required to produce LC3, viz. clinker, limestone, kaolinite clays and gypsum were identified in several states in India through visits and contacts at various locations with existing cement companies. While clinker, gypsum and limestone are easily available at all locations where cement is currently being produced, it was found that kaolinite clays can be easily found at least in the states of Rajasthan, Gujarat (western India), Kerala (southern India) and West Bengal (eastern India). Thermogravimetric analysis of various qualities of clays from Rajasthan and West Bengal showed that they potentially contained between 20% and 80% kaolinite. According to the Indian Bureau of Mines, these clays, commonly known as China Clays, are available in 22 states in India [8]. Due to logistical reasons of calcination, grinding and blending, it was finally decided to acquire the clays from mines in the state of West Bengal. Two clays, one with around 70% to 80% kaolinite content and another with around 20% to 30% kaolinite content were chosen for the study (Table 1). This kaolinite content was estimated on the basis of the chemical composition of the clays and the weight loss in the clays between 300°C and 600°C, which was further confirmed using XRD and FTIR measurements. Lime reactivity tests, carried out as described in IS1727-1967, on the clays calcined in a laboratory scale muffle furnace also confirmed that the first clay did have a high kaolinite content and the second clay had less kaolinite content.

Table 1. Properties of clays used in production

S.No.	Chemical Constituents	Clay 1	Clay 2
1	Silica (SiO_2)	43.30%	55.78%
2	Alumina (Al_2O_3)	36.35%	17.46%
3	Ferric Oxide (Fe_2O_3)	2.56%	8.89%
4	Calcium Oxide (CaO)	0.46%	4.84%
5	Loss on Ignition (LOI)	13.94%	9.49%
6	Colour	White	Red
7	Lime reactivity	9.25 MPa	1.38 MPa

Table 2. Properties of limestones used in the study

S.No.	Parameter	Limestone A	Limestone B
1	LOI	37.04%	35.1%
2	SiO_2	14.04%	18.2%
3	R_2O_3 (Alkali)	3.1%	1.1%
4	CaO	36.29%	42.84%
5	MgO	8.71%	1.08%

Two grades of limestone, one marginally lower than the cement grade and having a relatively high dolomite content and another of the cement grade were chosen for the study (Table 2). A chemical gypsum was obtained from a fertiliser plant in Haldia, Eastern India. Clinker obtained from a cement plant Bilaspur Chattisgarh was used for the production (Table 3).

3. CALCINATION OF CLAYS

It was found that although many facilities that calcine clays in rotary and vertical shaft kilns were available in India, the calcination of only a few tonnes of clay in

Table 3. Chemical analysis of clinker used in the production of LC3 (% of mass)

SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	Na_2O	K_2O	SO_3	P_2O_5	MnO	Cl	LOI
20.8	4.29	3.94	0.18	63.3	1.31	0.09	0.77	2.47	0.01	0.01	0.03	2.57

Table 4. Blends of LC3 produced

	LC3 A	LC3 B	LC3 C	LC3 D
Clay	Clay 1	Clay 1	Clay 2	Clay 2
Limetsone	Limestone A	Limestone B	Limestone A	Limestone B
Colour	Grey	Grey	Red	Red

these kilns was difficult due to their high production capacity. Furthermore, since these kilns usually calcine clays that are purer than the ones to be used in this study, they could contaminate the calciners thereby affecting their regular production. It was, therefore, decided to carry out static calcination of the clays in shuttle kilns used in ceramic production. An oil-fired shuttle kiln was used for the calcination. The clays were packed in burnt clay containers, called saggars, and fired at calcination temperatures. In each round, approximately 1.5 to 2 tonnes of clay was loaded into the furnace and thermocouples placed at several locations inside the furnace and inside the saggars. While earlier TGA measurements had shown that a sustained temperature of at least 600 °C was required for the complete calcination of the two clays, it was found that despite getting flame temperatures of 1100 °C, it was not possible to reach temperatures above 500 °C at the centre of the clay containers. Later laboratory measurements confirmed that approximately 50% of the clay was calcined, while the remaining 50% was not calcined. The calcined kaolinite content was later estimated to be approximately 40% and 10% respectively in clays 1 and 2.

4. BLENDING OF MATERIALS

The required materials were blended in a cement grinding unit in Kharagpur, West Bengal. Approximately 10 tonnes each of four blends of 50% clinker, 30% calcined clay, 15% crushed limestone and 5% gypsum were inter-ground in the ball mill of the grinding unit with a capacity of approximately 5 to 6 metric tonnes per hour to produce four blends of LC3 (Table 4). The limestone was fed as a fine powder and the clay and gypsum were fed as lumps. The clinker used to produce LC3 was also ground along with 5% gypsum to produce ordinary Portland cement (OPC) for comparison with the LC3 blends. The process of blending of each batch of LC3 was completed in approximately 2 hours. It must be noted here that although the blending process was found to be proper, it was not possible to obtain cements with uniform properties since the calcined clay was only partly calcined.

In order to avoid workability issues with the cements, it was decided to reduce the time of residence of the cements in the ball mill leading to a relatively coarse grinding of the cement. Still, a relatively high Blaine's fineness of the LC3 blends was obtained (Table 5) due to the 15% fine limestone content. A similar residence time was also kept for the OPC and the Blaine's fineness was found to be significantly lower.

5. LABORATORY TESTING OF CEMENTS

The blended cements were tested for their setting times and standardised strengths and the results were compared with the OPC produced using the same clinker and Portland pozzolanic cement (PPC) produced by blending the OPC with locally available fly ash. The standard consistency of the LC3 was found to be marginally higher than that of the OPC, probably due to the fine limestone used in the blends. The setting time of the LC3 blends, although lower than the OPC, was found to be within

Table 5. Properties of LC3 and OPC blends measured in laboratory

Property	LC3 A	LC3 B	LC3 C	LC3 D	OPC	PPC
Standard consistency (%)	32.5	33	34	34	29	29
Initial setting time (min.)	34	33	101	105	190	-
Blaine fineness (m^2/kg)	534	534	520	462	289	-
Mortar cube strength (MPa) (Water-cement ratio)	34.4 (0.445)	40.8 (0.45)	24.7 (0.46)	27.2 (0.46)	31.0 (0.43)	23.3 (0.43)

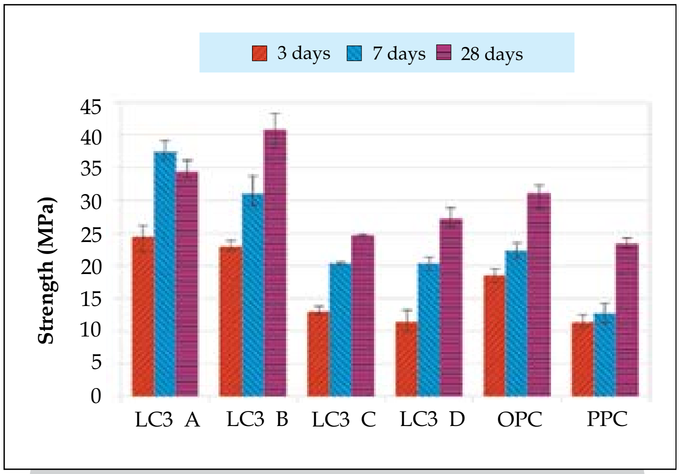

Figure 1. Comparison of strengths of mortars produced using LC3, OPC and PPC blends using water to cement ratios listed in Table 5

the requirements of the standards. It was found that the blends with higher quality clay set faster than the others.

The standardised mortar strengths of the LC3 blends with the low quality clay were found to be comparable to the OPC, while the strength of the blends with the higher quality clay gave strengths higher than the OPC, despite the higher water to cement ratios and the low clinker factors in the LC3 blends. It must be noted here that relatively low strengths were obtained due to the sub-optimal grinding of all the cements.

6. LABORATORY TESTING OF CONCRETES

Several concrete mixtures were designed and produced in the laboratory using all the blends of LC3, OPC and PPC. Polycarbonxylate Ether based chemical admixture was used to achieve a slump in the range of 75 mm to 100 mm,

Figure 2. Concrete cylinders using OPC, LC3 A, B, C and D (from left to right) cast in the lab

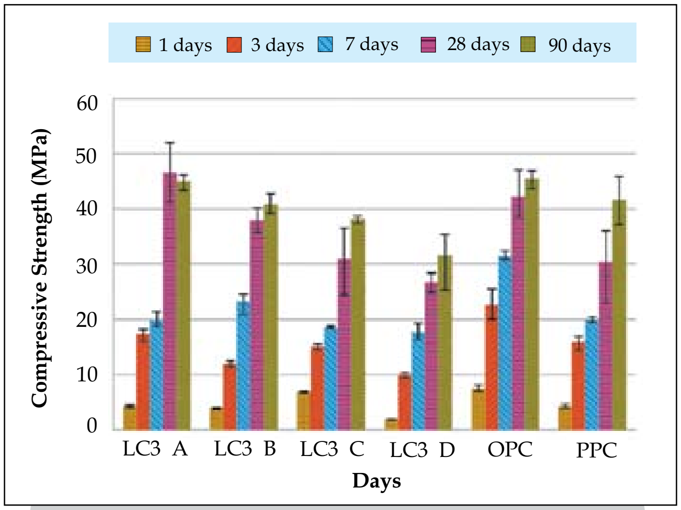

Figure 3. Comparison of strengths of concretes produced using LC3, OPC and PPC blends at 0.45 water to cement ratio

as the cements were found to give the best flow with LC3 blends. Perhaps due to the non-uniform calcination of the clays, a large batch-to-batch variation in the strengths of the concrete was observed, however, it was generally observed that for a given water to cement ratio, concretes using LC3 A gave the highest 28 day strengths, while the strengths from blend B was slightly lower. Strengths using LC3 C and D were similar to those obtained using the PPC. Figure 3 shows the variation of strengths of the concretes using a fixed water to cement ratio of 0.45.

7. FIELD TESTING AND PRODUCTION OF CONSTRUCTION MATERIALS

To test the cements and compare them with baseline PPC (PPC is the most used cement in normal building

Figure 4. MCR tiles produced using LC3 A, B, C and D (left to right)

materials) several types of building materials were produced in actual production scale. These were micro concrete roofing tiles, solid concrete bricks, hollow concrete blocks, RCC door and window frames and low duty paving blocks of various water: cement ratios. The tests of all the blends in actual production scale compared well with the laboratory results. Except blend B, all the blends had a similar water to cement ratio compared to Pozzolana Portland Cement. In blend B the water to cement ratio was required to be slightly increased in order to achieve the same workability and productivity as the PPC (Figure 5). Generally, the strengths obtained in all cases were equivalent to or higher than the products made using the PPC and trends similar to those obtained in the laboratory were observed. It must be noted here that the PPC used in this production was commercially obtained from Jhansi district and is not the same PPC as that used in sections 5 and 6 of this article.

The graph in Figure 6 shows the breaking load of 250 mm by 500 mm Micro Concrete Roofing (MCR) tiles in actual production scale compared to PPC and OPC. These loads were obtained through three-point bending tests with a span of 300 mm and the load in the middle.

During production, generally an MCR tile with PPC takes around 30-35 seconds to manufacture. For all the blended cements except Blend B, with same water to cement ratio, the vibration time was between 30-35 seconds. For Blend B, the water to cement ratio was increased to get the same production rate. Thus it can be concluded that the various types of blended cement produced will not affect the productivity of the building materials for equivalent or even better quality product.

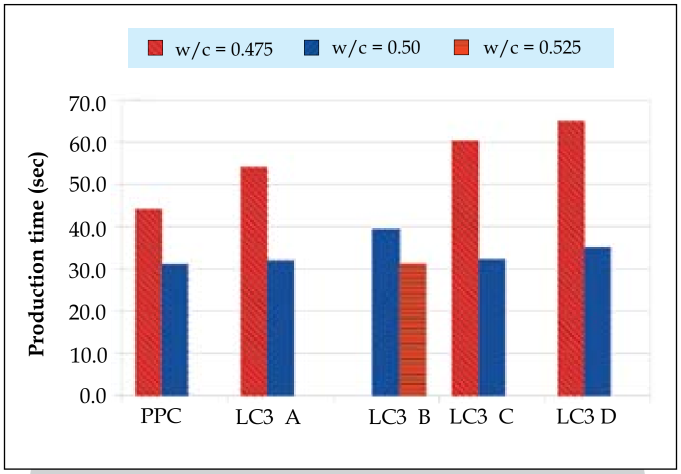

Figure 5. Comparison of time required to produce one MCR tile using various cements

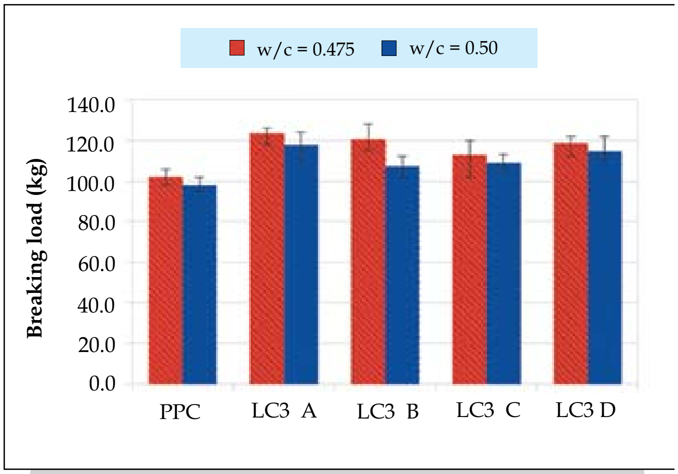

Figure 6. Comparison of the breaking load of MCR tiles using various cements measured using 3-point bending test with 300 mm distance between supports

The construction of a two-storey reinforced concrete building with hollow concrete block infills and micro-concrete roofing tiles made of all four blends of the LC3 is currently underway. The production of the blocks and tiles is nearing completion and the upper slab of the ground floor has been cast. No special processes or workers had to be used during the production of the blocks and tiles, and the construction process. This shows that unlike some of the other alternative cements, LC3 can be used in conventional construction without the need for specialised processes or training to workers.

Figure 7. Construction of two-storey house using LC3 blends in progress

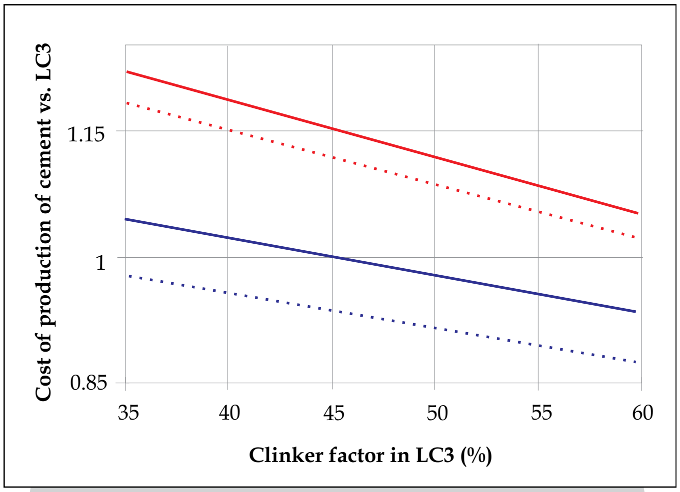

Figure 8. Relative cost of PPC and PSC vs LC3 for specific scenarios: Solid lines are for blends containing 40% fly ash and dashed lines for blends containing 50% slag. Red lines are for a scenario where fly ash or slag was available 250 km from the plant and clay was available 100 km from the plant. Blue lines are for scenario where fly ash or slag was available at a distance of 100 km from the plant and clay at a distance of 200 km from the plant.

8. PRELIMINARY ECONOMIC ANALYSIS

A preliminary analysis of the cost of production of LC3 with reference to the cost of production of OPC and PPC shows that it would be economical to produce LC3 in many situations, especially at locations where fly ash is not easily available. Even at locations where fly ash can be found near-by, it can be advantageous to produce LC3 due to the higher strength achieved in LC3 at lower clinker factors. Figure 8 shows the approximate relative cost of producing other types of cements vs. producing LC3 for different clinker factors used in LC3, assuming a similar cost of uncalcined clay, fly ash and slag, and the distance for which the slag, fly ash and calcined clay have to be transported. It can be seen that even LC3 blends with a clinker factor of 60% become economical when the fly ashes or slags are available 150 km farther than the clays. Furthermore, blends with 40% clinker factor can be economical even when the fly ash has to be transported 150 km less than the calcined clay.

9. SUMMARY AND CONCLUSIONS

A pilot production of LC3 blends was carried out using available, but non-optimal technologies in India, by personnel who are inexperienced in the production of cements. It was found that a complete calcination of the clay could not be carried out by the technology used and only half the clay could be calcined. Because of this, the blends of cements were also found to be non-uniform giving a high variation in the strength results. Furthermore, since the limestone was fed in a ground form, it significantly increased the fineness of the cement, with little contribution to its reactivity and strength development. Due to the added fineness, the cements had to be ground in a coarse form and relatively lower strengths were obtained in the mixes.

Despite the problems in the production of the LC3 blends, it was found that the blends that used the higher quality clays consistently gave better strengths that the OPC in mortars, concretes and building materials despite having less than 40% calcined kaolinite content in the calcined clay used. Furthermore, the blends that used a clay containing only around 10% calcined kaolinite achieved strengths similar to the PPC both in the laboratory and in the field. The results show that not only can LC3 provide us with a cement which has good early and long term strengths, but also that the LC3 technology is extremely robust and that even blends produced in extremely sub-optimal conditions can give good properties.

A preliminary analysis shows that LC3 can be produced economically with respect to OPC and PPC, especially at locations where there is a shortage of fly ash, but even otherwise due to the lower clinker factor that can be achieved in the blends.

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