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Research paper

Hydrated burnt clay–lime mixes: Effects of curing time and lime addition

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ABSTRACT

Effects of hydration time (up to 28 days) and lime addition (≤20 wt.%) on the changes of the microstructure of a kaolinitic–illitic clay heated at 600 °C were investigated using X-ray diffraction, scanning electron microscopy and Fourier transform infrared spectroscopy. Physical properties (strength, density, water absorption) of the cured blends were measured. The relations between properties and the operating factors were formulated using response surface methodology (RSM). Flocculation–agglomeration, carbonation and hydrate (C–S–H and CAH_{10}) formation were the main happening transformations. The pozzolanic reactions involved metakaolin and to a lesser extent illite. Based on RSM results, both factors had positive effects on strength and their interactions were synergistic. However, they manifested opposite effects and significant antagonistic interactions on density and water absorption.

1. Introduction

Blends of burnt clays and lime have been used as cementing materials before the discovery of ordinary Portland cement (OPC), and have received much attention recently (Sabir et al., 2001). As main constituents of clays, clay minerals (hydrated alumino-silicates) may behave as pozzolans. As a matter of fact, their pozzolanic activity enhances by thermal activation. Considering for instance metakaolin, which is a product of heated kaolinite, it reacts with lime and water forming thus cementitious hydrates such as C–S–H gel, C_2ASH_8 (strätlingite), C_4AH_{13} and C_3AH_6 (Siddique and Klaus, 2009).

Burnt clay based mortars could be convenient for the restoration of historical buildings (Budak et al., 2008). As compared to OPC mortars, they are inexpensive, environment-friendly materials and may present diverse practical advantages (Lea, 1974). However, their use depends on, among others, the chemical and mineralogical compositions of clays, grain size distribution, operating conditions and technical characteristics (drying shrinkage, water absorption, strength, etc.).

The pozzolanic behaviour of burnt clays, as compared to that of metakaolin, has been the subject of few investigations (Cara et al., 2006; Habert et al., 2009). In addition, much less attention has been paid to the pozzolanic behaviour of illite and metakaolin assemblage. On the other hand, few studies relevant to the effects of experimental factors on technical properties of cured burnt clay–lime mixes, using mathematical models, have been reported (Fortes-Revilla et al., 2006).

In this study, changes of the microstructure and some physical properties of hydrated mixes of a burnt kaolinitic–illitic clay and lime against ageing time and lime addition were investigated. The relations between properties and the later operating factors were established using response surface methodology (RSM), and the effects of factors were evaluated.

2. Materials and experimental procedures

The clay material was from a red argillaceous deposit at the valley of Ourika (High-Atlas Mountain, Morocco). Its mineralogical and chemical compositions are reported in Table 1. It was heated at 600 °C for 2 h in open atmosphere to transform kaolinite into metakaolin without altering illite (Hajjaji et al., 2002).

The lime was a locally manufactured material purchased from a common shop. Its mineralogical and chemical compositions, determined by X-ray diffraction and X-ray fluorescence spectroscopy respectively, are given in Table 1.

Samples of both materials were crushed and manually grounded, and oven dried at 105 °C. Their particle size distributions were quite similar and the grain size was <0.5 mm (Fig. 1). Density, water absorption and pH of the clay were 2.5 g/cm^3, 26% and 8 respectively.

Amounts of dried burnt clay and lime (Table 2) were introduced in a glass jar, and handily shook for about 10 min. The blends were sufficiently damped to prepare homogenised pastes. The ratio: water/blend approximated 40%. From the plastic mixtures, test-pieces ($5 \times 7.2 \times 10$ mm^3) were shaped using a steel mould, and aged 7, 14, 21 and 28 days within semi-closed humid boxes kept at room temperature (about 22 °C). For experiments, aged samples were dried at room temperature for 24 h and gradually heated in an oven-dryer until 105 °C, wherein they were kept for at least 3 h.

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Table 1
Mineralogy and chemical compositions of the studied clay and lime.

Mineralogy							
Clay				Lime			
Kaolinite	Illite	Quartz	Hematite	Portlandite	Lime	Calcite	
Chemical composition							
	SiO_2	Al_2O_3	Fe_2O_3	MgO	CaO	Na_2O	K_2O	LI[a]
Clay	43.2	23.3	9.1	5.1	1	0.8	5.7	6.8
Lime	5.8	1.9	0.2	2.1	89.3	Tr[b]	0.1	nd[c]

[a] Loss on ignition.
[b] Traces.
[c] Not determined.

Phase identification was performed by X-ray diffraction (XRD) using a Philips X'Pert MPD diffractometer operating with a copper anode (Kα radiation; λ = 1.5418 Å) and by Fourier transform infrared spectroscopy (FT-IR), using a Perkin Elmer spectrophotometer functioning in the range 4000 and 400 cm^{-1}. The microstructure was examined by a JEOL JMS 5500 scanning electron microscope apparatus, equipped with a Falcon EDAX system. For this goal, fragments of cured samples were coated with carbon.

Density (d) of aged samples was determined using a pycnometer and edible oil (d = 0.918 g/cm^3) as a solvent. The flexural strength (σ in MPa) of cured test-pieces was calculated according to the relation: $\sigma = 3/2\ [(F \times L) / (b \times h^2)]$, where F (in N) is the maximum load, L = 2.8 cm, 7.1 < h < 7.2 mm and 10.2 < b < 10.4 mm. F was measured with the three point method, using an EX 150 Deltalab apparatus manufactured by the Deltalab-SMT company (Carcassonne, France). The applied load is given by the deflection of a loading beam, calibrated at the factory, and the deflection of the specimen is measured on dial gauges.

For water absorption (WA) measurements, cured samples were placed on a water immersed sponge and periodically weighted. The amount of absorbed water was deduced from the saturation plateau of plotted kinetic curves. The standard deviations for WA, σ and d were 0.8%, 0.4 MPa and 0.03 g/cm^3 respectively.

3. Model presentation

The equation expressing the variation of a studied physical property (Y) of the aged samples as a function of curing time (t) and lime addition (τ) was established by the use of the response surface methodology, adopting the following polynomial model (Ferreira et al., 2003; Khalfaoui et al., 2006):

$$Y = a_0 + a_1X_1 + a_2X_2 + a_{11}X_1^{\ 2} + a_{22}X_2^{\ 2} + a_{12}X_1X_2.$$

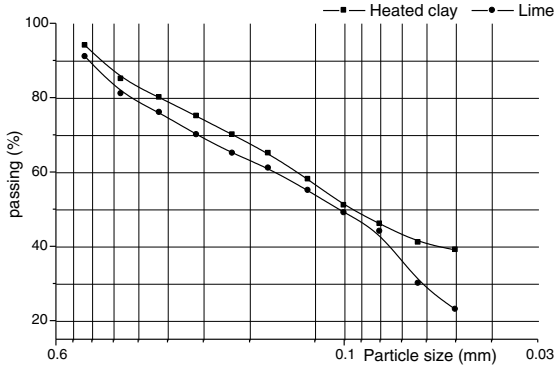

Fig. 1. Particle-size distribution of the used raw materials.

Table 2
Planned experimental conditions and measured values of the studied properties (Y_1: bending strength; Y_2: density; and Y_3: water absorption).

Run	X_1	X_2	t[a] (days)	τ[b] (wt.%)	Y_1 (MPa)	Y_2 (g/cm^3)	Y_3 (%)
1	−1.0000	−1.0000	7	2	0.335	2.460	–
2	−1.0000	−0.5555	7	6	0.960	2.420	24.03
3	−1.0000	−0.3333	7	8	1.070	2.450	26.02
4	−1.0000	0.1111	7	12	1.110	2.420	25.80
5	−1.0000	0.5555	7	16	0.587	2.510	30.64
6	−1.0000	1.0000	7	20	0.955	2.540	28.02
7	−0.3636	−1.0000	14	2	0.360	2.680	–
8	−0.3636	−0.5555	14	6	1.300	2.640	25.94
9	−0.3636	−0.3333	14	8	1.510	2.730	25.62
10	−0.3636	0.1111	14	12	3.710	2.760	23.75
11	−0.3636	0.5555	14	16	2.450	2.780	26.87
12	−0.3636	1.0000	14	20	2.568	2.810	25.56
13	0.2727	−1.0000	21	2	0.370	2.800	–
14	0.2727	−0.5555	21	6	2.030	2.800	24.37
15	0.2727	−0.3333	21	8	2.330	2.930	25.31
16	0.2727	0.1111	21	12	2.583	2.850	25.24
17	0.2727	0.5555	21	16	2.273	2.850	26.36
18	0.2727	1.0000	21	20	3.278	2.860	26.50
19	0.9091	−1.0000	28	2	0.406	2.950	–
20	0.9091	−0.5555	28	6	2.200	2.850	23.25
21	0.9091	−0.3333	28	8	2.842	2.760	22.91
22	0.9091	0.1111	28	12	2.900	2.690	22.62
23	0.9091	0.5555	28	16	3.164	2.640	24.00
24	0.9091	1.0000	28	20	4.100	2.630	19.69

[a] Curing time.
[b] Lime content.

X_1, X_2 are the coded variables related to the factors t and τ: $X_1 = (t - t_o) / \Delta t$; $X_2 = (\tau - \tau_o) / \Delta\tau$; where t_o = 18 days, Δt = 11 days, τ_o = 11 wt.% and Δτ = 9 wt.%. t_o and τ_o are the values at the centres of the investigated domains of t and τ. Δt and Δτ are the variations steps of the variables t and τ.

a_0 is a constant; a_1 and a_2 are coefficients representing the weights of the effects of the curing time and lime addition respectively; a_{12} is a constant expressing the interaction effect between the curing time and lime addition; and a_{11} and a_{22} can be regarded as the curve shape parameters. a_i, a_{ii} and a_{ij} were calculated by the least square regression using the software NEMROD (new efficient methodology for research using optimal design) (Mathieu and Phan Tan Luu, 1980). The experimental values of the studied properties (bending strength, density and water absorption), used for the calculation of the model coefficients, are reported in Table 2.

The significance and the validity of the model were evaluated by the analysis of variance (ANOVA). The statistical calculations were performed by the NEMROD software.

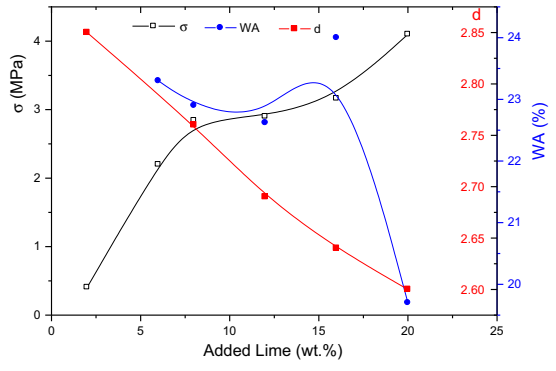

Fig. 2. Variations of the bending strength (σ), density (d) and water adsorption (WA) of hydrated mixes (28 days) against lime addition.

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