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INFLUENCE OF CHEMICAL AND MINERALOGICAL COMPOSITION OF METAKAOLIN ON MORTAR CHARACTERISTICS

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ABSTRACT
Three different metakaolins processed by a Portuguese factory were studied aiming to assess the influence of chemical and mineralogical composition of metakaolin on mortar characteristics. Mineralogical analysis of both the fine and clay fractions was carried out by X-ray diffraction. Chemical composition (major elements) was analyzed by X-ray fluorescence. Mineralogical composition reveals some significant differences between the studied samples, in both analyzed fractions, metakaolin 2 being richer in kaolinitic minerals. From a chemical point of view, siliceous content is always high (around 60 %) as well as Al_2O_3 content (around 30 %); once again metakaolin 2 is the one richer in Al_2O_3 and poorer in sodium and potassium. Metakaolin from the three different batches was incorporated in lime mortars in order to produce a pozzolanic reaction and hence enable them to harden in high relative humidity conditions or, when access to CO_2 is limited, as in the case of mortars supporting glazed tiles. Mortars with the same volumetric ratio were tested for flexural and compressive strength at ages of 28 and 90 days, and the module of elasticity was determined. A relation between the mechanical strength achieved by lime mortars with the addition of metakaolin from different batches and metakaolin mineral and chemical composition was assessed. The knowledge of the influence of the metakaolin composition on the strength of the mortars is an important step in improving their performance and extending their application.

KEYWORDS: metakaolin, mineralogical, chemical composition, mortar

1. INTRODUCTION

The addition of pozzolanic materials improves the mechanical characteristics of these lime mortars and contributes towards a higher durability.

Metakaolin is an artificial pozzolan obtained by the calcination of kaolinitic clays over a specific temperature range. It is a material with pozzolanic properties and can be added to lime mortar mixes to provide improved mechanical and water behaviour characteristics for use in conservation mortars. For this particular application cement mortars are inadequate, due to their excessive brittleness, low plasticity and high elastic modulus and high content in soluble salts (Teutónico et al., 1994; Moropoulou et al., 2005).

A further advantage of lime/pozzolan mortars is their lower environmental impact, when compared to cement mortars, due to lower energy consumption during production and CO_2 absorption by carbonation. The addition of metakaolin to mortars and concretes also has a positive effect in terms of durability (Siddique and Klaus, 2009; Silva, 2005).

In Portugal, metakaolin is still used on a small scale, whereas in some countries (e.g. the U.K. and USA) metakaolin is commercialized and is more widely used. Knowledge of the influence of the metakaolin composition on the properties of metakaolin-lime mortars is an important step in improving their performance and extending their application in the field of conservation.

Kaolinitic clays are available in Portugal, especially in the north and centre of the country (Ferraz, 2004; Gomes, 1990), although many quarries are no longer active due to lack of demand. However, a growing scientific interest in the use of metakaolin in mortars and concretes, in order to improve mechanical strength or reduce alkali-silica reaction (Fortes-Revilla et al., 2006; Kim, 2007; Ramochlan, 2000; Silva, 2005), together with the prospective lack of traditional pozzolanic materials such as fly ash and silica fume, are inducing the industrial sector towards metakaolin production.

Metakaolin is obtained from the calcination of kaolinitic clays at temperatures in the range of 700-800 ºC (Badogiannis, 2005; Sabir, 2001), high enough to allow for loss of hydroxyls but below temperatures that cause the formation of a vitreous phase and crystallization of other phases such as mullite.

Requisites for conservation mortars comprise a low elastic modulus, sufficient flexural strength and adequate behaviour in terms of water intake and drying (Veiga, 1998; Veiga et al, 2007). Ductility is also an important property that may be evaluated using a restrained shrinkage test, especially developed for rendering mortars (Veiga et al, 2007). Chemically, these materials must also guarantee compatibility issues.

At present, building conservation practice uses air lime or hydraulic lime as preferable binders. However air lime presents certain problems including slow setting, inability to harden under water, lack of durability and specific application procedures whilst hydraulic limes of national production have proved inadequate due to difficulty in drying and irregular mechanical characteristics.

The use of pozzolans in air lime mortars is a subject of recent studies (Fortes-Revilla et al., 2006; Velosa, 2006) and results suggest that, in the appropriate proportions, they produce an increase in mechanical strength and durability of mortars, meeting water intake and drying requirements. Additionally, these mortars have low cracking susceptibility (Veiga et al., 2007), a factor of major importance, crucial in terms of efficiency towards limiting water absorption of walls and increasing durability.

The use of air lime and metakaolin in mortars may also contribute towards sustainability, due to lower temperatures used in the production process, when compared to other binders. Also air lime mortars have the additional capacity to re-absorb CO_2 from the atmosphere during the carbonation process that leads to mortar hardening (Holmes and Wingate, 1997).

Two different metakaolins processed by a Portuguese factory were studied, compared to an commercialized one, aiming to assess the influence of chemical and mineralogical composition of metakaolin on mortar characteristics.

2. MATERIALS, MORTAR COMPOSITIONS AND CONDITIONING

Mortars were formulated with powdered commercial air-lime (CL 90, containing over 90 % CaO), a siliceous river sand mainly composed of quartz and three different metakaolin samples (K1, K2 and K3) from the same producer but with differences relating to source and production technique. The source of kaolin extraction is a unique quarry that has various strata. The raw material for metakaolin production was removed from different areas of the quarry and at different depths. The production procedure is being improved with an adjustment of calcination temperature, improvement of the grinding process and an increase in homogenization of the final product. The first output was metakaolin K1 and the last output, after improvements, was metakaolin K3.

These products were characterized by X-ray diffraction (XRD) using Philips X-Pert Pro X-Ray Diffractometer to identify crystalline phases. Mineralogical analysis of both, the fine (<63 µm, randomly oriented powder samples) and clay (< 2 µm, randomly oriented powder samples and oriented aggregates) fractions was carried out according to Moore and Reynolds (1997), Galhano et al. (1999) and Oliveira et al. (2002).

Chemical composition (major elements) was analyzed using Philips PW 1400 X-Ray Fluorescence Spectrometer. Metakaolin samples were prepared as for XRD. Together with XRD, this analysis provides complementary information on metakaolin composition.

Mortar proportions (lime:pozzolan:sand) were prepared by volume and added water was calculated in relation to required consistency, (i.e. similar flow values of around 130), which corresponds to appropriate workability for this type of mortar. A lime mortar with a lime/sand volumetric ratio of 1:3 was used as comparative mortar and a ratio 1:0.5:2.5 (lime: pozzolan: sand) was used for the test mortars taking into account that the pozzolanic material itself is not cementitious but acts as a binder by reacting with and consuming lime (Moropoulou et al., 2004; Sabir et al., 2001). A 1:3 cement mortar was also used for comparison although its characteristics make it inadequate for use in building conservation.

Mortar specimens were prepared and conditioned in a climatic chamber following standard NP EN1015-11: Methods of test for mortar for masonry - Part 11: Determination of flexural and compressive strength of hardened mortar. Specimens were stored for the first 7 days at 20±2 ºC in a plastic bag. They were then maintained at the same temperature, but at a relative humidity of 65±5 %. Mortars with no pozzolanic addition (L) were not stored initially in a plastic bag, as previous attempts to implement this, by the authors and by other researchers, resulted in a retarded hardening of pure lime mortars, possibly due to difficulty in carbonating in such conditions.

Compressive and flexural strength testing was performed following standard NP EN1015-11: Methods of test for mortar for masonry - Part 11: Determination of flexural and compressive strength of hardened mortar. The dynamic modulus of elasticity was determined following Re-port LNEC 427/05-NRI, based on the determination of the vibration resonance frequency.

3. RESULTS AND DISCUSSION

Mineralogical composition reveals some significant differences between the studied samples, in both analyzed fractions; concerning fine fractions, metakaolin 1 (K1) is the one richer in quartz whereas metakaolin 2 (K2) is richer in kaolinitic minerals; concerning clay fractions, metakaolin 1 shows clear

Table 1 XRF results of metakaolin samples.

	SiO_2 (%)	Al_2O_3 (%)	Fe_2O_3 (%)	MgO (%)	CaO (%)	Na_2O (%)	K_2O (%)	TiO_2 (%)	LOI (%)
K 1	62.62	28.63	1.07	0.15	0.06	1.57	3.46	0.36	2.00
K 2	59.90	32.29	1.28	0.17	0.04	0.24	2.83	0.36	2.80
K 3	62.48	28.72	1.01	0.13	0.03	2.45	3.55	0.34	1.20

Table 2 Pozzolanic reactivity of metakaolin samples.

Sample	OH^- concentration (mmoles/l)	CaO concentration (mmoles/l)
K 1	38.80	7.19
K 2	43.20	7.70
K 3	52.19	5.56

evidences of disordered kaolinite whereas 3 shows a more ordered kaolinite as well as some evidences of dickite, metakaolin 2 showing an intermediate composition.

From a chemical point of view (Table 1), siliceous content is always high (around 60 %) as well as Al_2O_3 content (around 30 %), but once again the three samples show some clear differences; K2 is the one richer in Al_2O_3 being also poorer in alkalies (sodium and potassium); K1 metakaolin shows the oposite characteristics; K3 metakaolin shows a composition similar to K1.

Metakaolin from three different batches was incorporated in lime mortars in order to produce a pozzolanic reaction and hence enable them to harden in high relative humidity conditions or, when access to CO_2 is limited, as in the case of mortars supporting glazed tiles.

Pozzolanic reactivity of metakaolin was measured following NP EN 196-5: Methods of testing cement, pozzolanicity test for pozzolanic cement. Although this testing method is usually applied to pozzolanic cements, it provided the most accurate results compared with other available methods when applied to various natural and artificial pozzolans (Velosa, 2006). The procedure consists of mixing given quantities of cement and pozzolan, that are left in an aqueous solution at constant temperature for 7 to 15 days and of the measurement, by titration, of $Ca(OH)_2$ remaining in the solution. Lower concentrations of OH ion and CaO indicate higher pozzolanic reactivity due to $Ca(OH)_2$ consumption by pozzolanic reaction. Results of tests on all metakaolin batches revealed clear pozzolanic reactivity of all metakaolin samples (Table 2). Mortars were monitored using SEM/EDS.

The products of pozzolanic reaction were clearly evident as seen in Figure 1 showing MK1 at the age of 28 days.

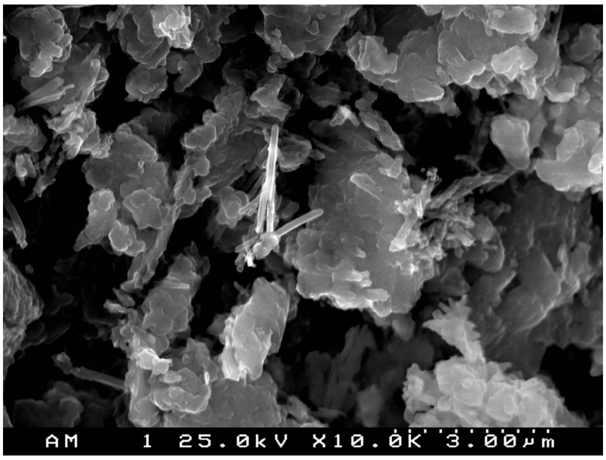

Fig. 1 Evidence of pozzolanic reaction (CSH formation) in mortar MK1.

Results of the flexural strength test (Figure 2) indicate an increase in strength of mortars MK2 and MK3 in relation to lime mortar with no addition. However, results of MK1, were very similar to those of lime mortar, probably due to the fact that K1 was from the initial batch that was produced at a lower temperature. The higher flexural strength displayed by mortars MK2 and MK3 is probably due to the pozzolanic action of metakaolin as the increase of mechanical behaviour of lime mortars due to the addition of pozzolans. It is promoted either by an increase in the compaction of mortar, with a special impact on compressive strength, or by the effect of pozzolanic reaction (Velosa, 2006).

Compressive strength results (Figure 3) show similarities with those of flexural strength, with a clear strength increase in mortars MK2 and MK3 in relation to mortar L, with no metakaolin, whilst mortar MK1 reveals an early strength increase in relation to mortar L, but attained strength stabilizes

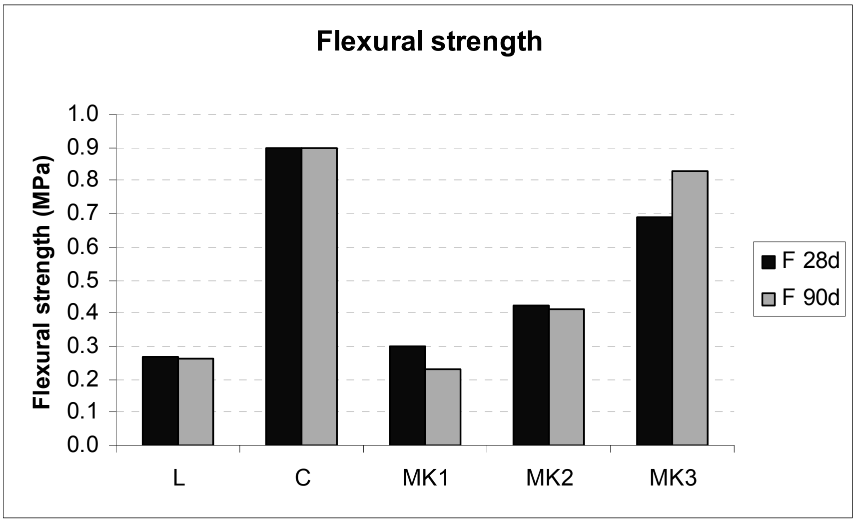

Fig. 2 Flexural strength of lime/metakaolin mortars (MK) compared with lime mortar (L) and cement mortar (C).

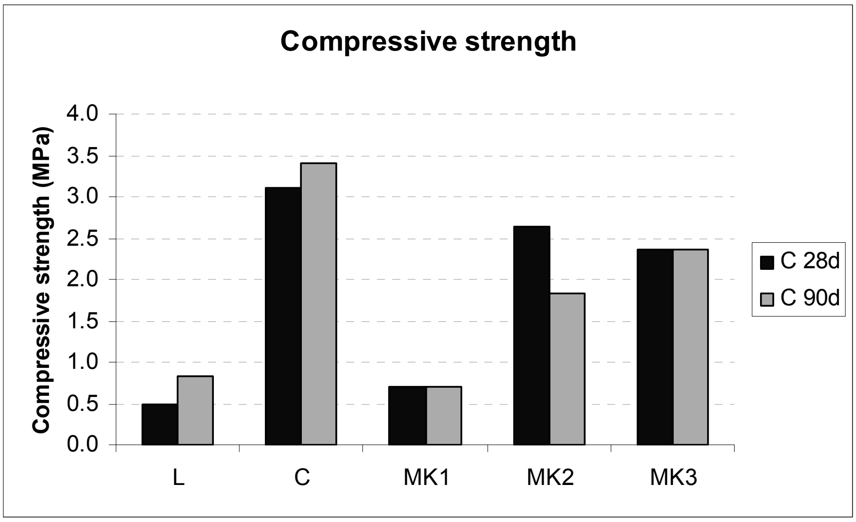

Fig. 3 Compressive strength of lime/metakaolin mortars (MK) compared with lime mortar (L) and cement mortar (C).

until the age of 90 days. At this age, compressive strength of MK1 is lower than that of the compared lime mortar. Reasons for lower strength of MK1 may be due to the fact that it wasn't fired at a high enough temperature to enable it to attain sufficient pozzolanic reactivity. The decrease in compressive strength from the age of 28 days to the age of 90 days in the case of MK2 is a phenomenon that has been observed in other mortars, namely those containing pozzolans; reasons for this are unclear but may be linked with microcracking due to shrinkage, to which flexural strength is very sensitive (Veiga and Carvalho, 1998).

Both in terms of flexural and compressive strength results, lime/metakaolin mortars reveal lower mechanical strength than the cement mortar used for comparison.

Elastic module of almost all tested mortars (Figure 4) with a lime binder is low, as desired, ranging from 2000MPa to 3000MPa, and variations from 28 days to 90 days are not significant. This is a fundamental property towards the judgement of application possibilities as renders with high elastic modules have low deformation capability and are therefore unsuitable for use in conservation work. Cement mortar displayed a relatively high elastic module, ranging between 5500MPa and 6600MPa and mortar MK3 also revealed a high value of almost 5000MPa at 90 days.

Although it is expected that pozzolanic additions increase mechanical strength of mortars, this did not occur with mortar MK1. A possible explanation for this is that K1 acted partially as an aggregate due to

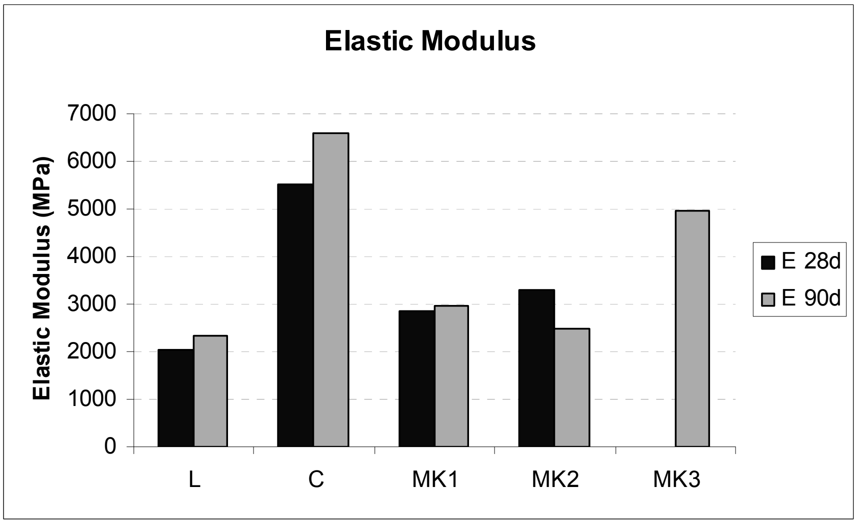

Fig. 4 Elastic module of lime/metakaolin mortars (MK) compared with lime mortar (L) and cement mortar (C).

lower firing temperature or inadequte grinding during production. Although all metakaolins reveal positive pozzolanic reactivity following standard NP EN 1015-11 (Table 2), this is a measure for initial pozzolanic reaction that continues to develop with time. Mechanical strength results at ages 28 and 90 days reveal that there is a possibility for unsatisfactory development of this reaction, as it has an immediate impact in compressive strength. The obtained results were consistent, revealing an increase in mechanical strength of lime mortars with pozzolanic additions in all mortars except for MK1.

4. CONCLUSIONS

1. The addition of pozzolanic materials increases the mechanical characteristics of these mortars. Mortars with the same volumetric ratio were tested in terms of flexural and compressive strength at ages of 28 and 90 days, and the module of elasticity was determined. Although generally metakaolin additions improved mechanical strength of the lime mortars, this did not occur with the use of metakaolin K1. All lime/metakaolin mortars had lower strength than an equivalent cement mortar. This fact, linked to chemical compatibility makes them more adequate for use in building conservation work.
2. A relation between the mechanical strength, achieved by lime mortars with the addition of metakaolin from different batches, and metakaolin mineralogical and chemical composition was assessed. Mortars using metakaolins richer in well ordered kaolinite and richer in Al_2O_3 (being also poorer in alkalies) showed better mechanical results.
3. The type of metakaolin employed is relevant, as metakaolin from different batches provides different results due to differences in the production process and raw materials. In the studied case, when K1 metakaolin was used, the mortars attained similar results to lime mortars. This may be due to the fact that K1 is richer in quartz and has a lower pozzolanic reactivity. However, after the improvement of the production process, by using an adequate calcination temperature, mortars using K2 and K3 gave better mechanical results due to their improved quality and inherent greater pozzolanic reactivity.

Further studies, taking into account cracking susceptibility and durability (climate and salts) will be undertaken. However, following these results and after in-situ tests had been performed, mortars with metakaolin (K2) were applied on a seaside fortress in Portugal, subjected to a harsh climate, with positive results so far (Veiga et al, 2009).

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REFERENCES

Badogiannis, E. et al.: 2005, Metakaolin as a main cement constituent. Exploitation of poor Greek kaolins. Cement & Concrete Composites, 27, 171–181.

Kim, H., Lee, S. and Moon, H.: 2007, Strength properties and durability aspects of high strength concrete using Korean metakaolin, Construction and Building Materials. 21 (6), 1229–1237.

Ferraz, E.J.: 2004, Kaolins of Alvarães: properties and ceramic applications. PhD Thesis, University of Aveiro (in Portuguese).

Fortes-Revilla, C., Martínez-Ramírez, S. and Blanco-Varela, M.T.: 2006, Modelling of slaked lime–metakaolin mortar engineering characteristics in terms of process variables, Cement and Concrete Composites. 28 (5), 458–467.

Galhano, C., Rocha, F. and Gomes, C.: 1999, Geostatistical analysis of the influence of textural, mineralogical and geochemical parameters on the geotechnical behaviour of the "Argilas de Aveiro" formation (Portugal). Clay Minerals, 34, 109–116.

Gomes, C.F. and Velho, J.A.G.: 1990, Kaolin deposits of Portugal. Geociências, Revista da Universidade de Aveiro, 5 (2), 27–36.

Holmes, S. and Wingate, M.: 1997, Building with Lime. Intermediate Technology Publications., London, UK

Moore, D.M. and Reynolds, R.C. Jr.: 1997, X-ray diffraction and the identification and analysis of clay minerals. Oxford New York, 2nd Edition, 378 pp.

Moropoulou, A., Bakolas, A. and Aggelakopoulou, E.: 2004, Evaluation of pozzolanic activity of natural and artificial pozzolans by thermal analysis. Thermochimica Acta, 420, 135–140.

Moropoulou, A., Bakolas, A., Moundoulas, P., Aggelakopoulou, E.: 2005, Reverse engineering: a proper methodology for compatible restoration mortars. In press, Proceedings of Workshop Repair Mortars for Historic Masonry, TC RMH. Delft: RILEM.

Oliveira, A., Rocha, F., Rodrigues, A., Jouanneau, J., Dias, A., Weber, O. and Gomes, C.: 2002, Clay minerals from the sedimentary cover from the Northwest Iberian shelf. Progress in Oceanography, 52, 233–247.

Ramlochan, T., Thomas, M. and Gruber, K.: 2000, The effect of metakaolin on alkali–silica reaction in concrete, Cement and Concrete Research. 30(3), 339–344.

Sabir, B.B., Wild, S. and Bai, J.: 2001, Metakaolin and calcined clays as pozzolans for concrete: a review. Cement & Concrete Composites, 23, 441–454.

Siddique, R. and Klaus, J.: 2009, Influence of metakaolin on the properties of mortar and concrete: a review. Applied Clay Science, 43 (3-4), 392–400

Silva, A.S.: 2005, Concrete degradation by alkali-silica reactions. Use of fly ash and metakaolin for its prevention. PhD Thesis (in Portuguese).

Teutonico, J.M., McCaig, I., Burns, C. and Ashurst, J.: 1994, The Smeaton Project: Factors affecting the properties of lime-based mortars. Bulletin of the Association for Preservation Technology, 25 (3-4), 32–49.

Veiga, M.R. and Carvalho, F.: 1998, Some performance characteristics of lime mortars for use on rendering and repointing of ancient buildings. Proc. 5th International Masonry Conference. London, 107–111

Veiga, M.R, Velosa, A. and Magalhães, A.C.: 2007, Evaluation of mechanical compatibility of renders to apply on old walls based on a restrained shrinkage test. Materials and Structures, 40 (10), 1115–1126

Veiga, M.R., Velosa, A. and Magalhães, A.: 2009, Experimental applications of mortars with pozzolanic additions: characterization and performance evaluation, Construction and Building Materials, 23 (1), 318–327

Velosa, A.L.: 2006, Lime mortars with pozzolans for use as ancient wall renders. PhD Thesis, University of Aveiro.