Investigations about the effect of aggregates on strength and microstructure of geopolymeric mine waste mud binders

Fernando Pacheco-Torgal [a,*], João Castro-Gomes [b,1], Said Jalali [c,2]

[a] *Castelo Branco Polytechnic Institute, Department of Civil Engineering, 6000 Castelo Branco, Portugal*
[b] *University of Beira Interior, Department of Civil Engineering, 6200 Covilhã, Portugal*
[c] *University of Minho, Department of Civil Engineering, 4800 Guimarães, Portugal*

Abstract

Geopolymeric binders appear to be an alternative to traditional Portland cement, due to high mechanical performances and environmental advantages. Some aspects related to the effect of aggregates in the microstructure and mechanical behaviour of geopolymeric mine waste mud (GMWM) binders are reported in the present study. Compressive and tensile strength of mine waste mud binders were analyzed. The factors investigated were the aggregate/binder ratio, the aggregate dimension and aggregate type, schist, granite and limestone.

Test results showed that GMWM binders have a very high strength at early ages and also possess a very high tensile strength. It's suggested that behaviour may be due to the dissolution of quartz and alumina in the presence of alkalis enhancing bonding between paste and aggregates.

The aggregate dimension showed only significant effect on tensile strength. Limestone aggregates showed a chemical bond to the alkali-activated paste but presented higher shrinkage. It was also found that no traditional porous ITZ was detected in GMWM binders.
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1. Introduction

Portland cement is the most used material in the worldwide construction industry and has a high level of CO_2 (1 ton of cement generates 1 ton of CO_2) and also its use tends to become less competitive compared to alternative ecological new binders like geopolymers [1]. Although research in this field has been published as "alkali-activated cement, or "alkaline cement" the term "geopolymer" is the generally accepted name for this technology. Geopolymerisation involves a chemical reaction between various aluminosilicate oxides with silicates under highly alkaline conditions, yielding polymeric Si–O–Al–O bonds indicating that any Si–Al materials could become sources of geopolymerization. Over the last years several authors have reported research in a large number of aspects related to geopolymeric based binders such as: dependence of the nature of source materials (geopolymers synthesised from calcined sources show a higher compressive strength than from raw materials) [2–4], immobilization of toxic metals [5,6], reaction mechanisms and hydration products [7–11], the role of calcium in geopolymerization [12–15], manufacture operations [16,17] and even the development of lightweight building materials [18].

There's however little research published about the effect of aggregates in geopolymeric materials. Some authors demonstrated that geopolymeric mortars had little differences from concrete synthesised from the same geopolymeric binder when low alkali mixtures were used [19], which is a surprising result because in OPC systems compressive strength increased through the addition of aggregates [20], although some influence in OPC systems has been reported by others [21,22]. In addition unlike OPC systems no apparent transition zone could be identified near the aggregates [23].

* Corresponding author. Tel.: +351 272 339300; fax: +351 272 339399.
E-mail addresses: fernandotorgal@est.ipcb.pt (F. Pacheco-Torgal),
castro.gomes@ubi.pt (J. Castro-Gomes), said@civil.uminho.pt (S. Jalali).
[1] Tel.: +351 275 329990; fax: +351 275 329969.
[2] Tel.: +351 253 510200; fax: +351 253 510213.

It's still not clear how aggregates behave in highly alkaline binders and if their size and absorption could influence that behaviour. Therefore, in this study the effect of some aggregates in the microstructure and mechanical behaviour of alkali-activated mine waste mud pastes and mortars is investigated.

2. Experimental program

2.1. Materials

Tungsten mine waste mud used in this study was subjected to a thermal treatment at 950 °C during 2 h, mineralogical composition and thermal conditions were described elsewhere [24,25].The XRD patterns indicated that mine waste mud consists mainly of muscovite and quartz which were identified by their characteristic patterns as follows: muscovite (card 46-1409) and quartz (card 46-1045). For those thermal conditions XRD patterns indicated that dehydroxylation did not result in a complete collapse of muscovite structure. Calcination leads to formation of an amorphous phase, causing an increase in the general background (BG) of XRD patterns and dominantly taking place in the calcinations interval from 850 to 950 °C, with a thermal behaviour similar to other phyllosilicate clay minerals [26]. The main muscovite peak ($2\theta = 8.8°$) persisted even after the sample had been heated at 950 °C although it decreased considerably. Peak area measurements revealed that about 12% of muscovite survived calcination at 950 °C. Molecular changes during dehydroxylation were also examined with infrared emission spectra (FTIR), confirming decrease in the absorption peaks at 3600–3700 (OH stretch), however the main muscovite peak did not disappear totally indicating only a partial transformation.

Compressive strength data related to alkali-activated mortars made with, respectively raw waste mud and calcined waste mud showed an increase of more than 300% (see Table 1) justifying the thermal treatment, besides mortars made with raw waste mud weren't very stable from a structural view point because they easily showed a lot of efflorescences in the surface of the specimens, due to non-reacted sodium ions which will react with atmospheric carbon dioxide leading to the formation of sodium carbonate crystals. Although calcination of mine waste mud in a static furnace, as it happens in this study may be expensive, some authors had already developed flash calciner industrial 800 Kg/h production units, capable of reduce calcination time to a few seconds and with the additional advantage of no further grinding operations [27,28] which will cut down the cost of down mine waste mud thermal treatment.

Table 1
Compressive strength of alkali-activated mine waste mud mortars

Curing (days)	Compressive strength (MPa)	
	Raw mine waste	Waste calcined at 950° during 2 h
7	8.4 (1.2)	28.4 (1.8)
14	9.3 (4.3)	37.0 (1.1)
28	11.2 (1.8)	39.6 (1.8)

Bracketed values are the standard deviation.

Table 2
Chemical composition and specific surface

Constituents (%)	Mine waste mud
SiO_2	53.48
Al_2O_3	16.66
Fe_2O_3	12.33
K_2O	7.65
Na_2O	0.62
MgO	1.27
SO_4	3.10
TiO_2	1.39
As	1.28
Other minor oxides	2.22
Blaine fineness (m^2/Kg)	357

The chemical composition and specific surface of the mine waste mud are shown in Table 2. Table 3 shows gradation and physical characteristics of aggregates. Limestone and granitic coarse aggregates have the same Faury maximum dimension (9.52) and similar fineness modulus respectively 5.5 and 5.2. Schist sand has a lower maximum dimension (2.78) and a fineness modulus of 2.38.

2.2. Mix proportion, strength testing and microanalysis of the binders

The samples composition is shown in Table 4. In this investigation the mortar was a mixture of aggregates, waste mud, calcium hydroxide, alkaline silicate solution and water. The mass ratio of mine waste mud: activator was 1:1. Calcium hydroxide was used with a percentage substitution of 10%, because it was found that percentage leads to the highest compressive strengths. An activator with sodium hydroxide (24 M) and sodium silicate solution ($Na_2O = 8.6\%$, $SiO_2 = 27.8\%$, $Al_2O_3 = 0.4\%$ and water = 63.2%) was used with a mass ratio of 1:2.5. Previous investigations showed that this ratio leads to the highest compressive strength results in alkali-activated mine waste mud mortars [29]. Distilled water was used to dissolve the sodium hydroxide flakes to avoid the effect

Table 3
Gradation and physical characteristics of aggregates

	Cumulative percentage retained		
	Schist (SC)	Limestone (LS)	Granite (GR)
	Sand	Coarse aggregate	
9.5 mm	0	0	0
4.76 mm	0	62.9	35.4
2.38 mm	0.9	89.1	91.5
1.19 mm	28.3	99.3	97.2
590 μm	62.2	99.4	98.5
297 μm	88.8	99.4	99.2
149 μm	98.7	99.7	99.4
74 μm	99.8	99.8	99.6
Dust content (<74 μm)	0.2	0.2	0.4
Fineness modulus	2.78	5.5	5.2
Max. dimension (mm)	2.38	9.52	9.52
Water absorption (%)	0.9	0.8	0.4
Density (Kg/m³)	2703	2659	2689

of unknown contaminants in the mixing water. The alkaline activator was prepared prior to use. The sand, mine waste mud and calcium hydroxide were dry mixed before being added to the activator, which according to Teixeira-Pinto et al. [16] is the mixing option that leads to the best results. To produce a workable mix extra water has been added as stated by Jahanian and Rostami [30]. The mass ratio of water/dry solid binder content was 3.6% in most of the samples, except for samples with an aggregate/binder mass ratio of 1.5 or 1.7 in those cases, the extra water percentages were respectively 7 and 10%. The sample composition is shown in Table 3.

Compressive strength data was obtained using $50 \times 50 \times 50$ mm^3 cubic specimens according to ASTM C109 [31]. The fresh mortar was cast and allowed to set at room temperature for 24 h before being removed from the moulds and kept at room temperature until tested in compression. Compressive strength for each mixture was obtained from an average of 3 cubic specimens determined at the age of 1, 3, 7, 14 and 28 days of curing.

Tensile strength was analysed with a flexural test. Specimens with $40 \times 40 \times 160$ mm^3 were used according to EN 1015-11 [32]. Tensile strength for each mixture was obtained from an average of 2 specimens determined at the age of 1, 3, 7, 14 and 28 days of curing.

For examination by scanning electron microscopy (SEM) mortar samples were embedded into a low viscosity epoxy resin, cut and polished. After the lapping stage an additional epoxy impregnation was performed, and the samples were relapped in order to ensure full resin impregnation and a good quality polish. The samples were then carbon-coated. A Hitachi S2700 scanning electron microscopy was used equipped with a solid backscattered detector and a Rontec energy dispersive (EDS) X-ray microanalysis system.

3. Results and discussion

3.1. Chemical composition and specific surface

The figures in Table 1 clearly show that mine waste mud consists essentially of silica and alumina, contaminated with arsenic and sulfur and with a high content of iron and potassium oxide. Although it's known that iron oxide contributes to the strength of Portland cement it is still an open question if iron also contributes to geopolymerization [2].

The SiO_2/Al_2O_3 atomic ratio is 5.5 higher than the one suggested by Davidovits of about 2 for making cement and

Table 4
Samples composition (in mass)

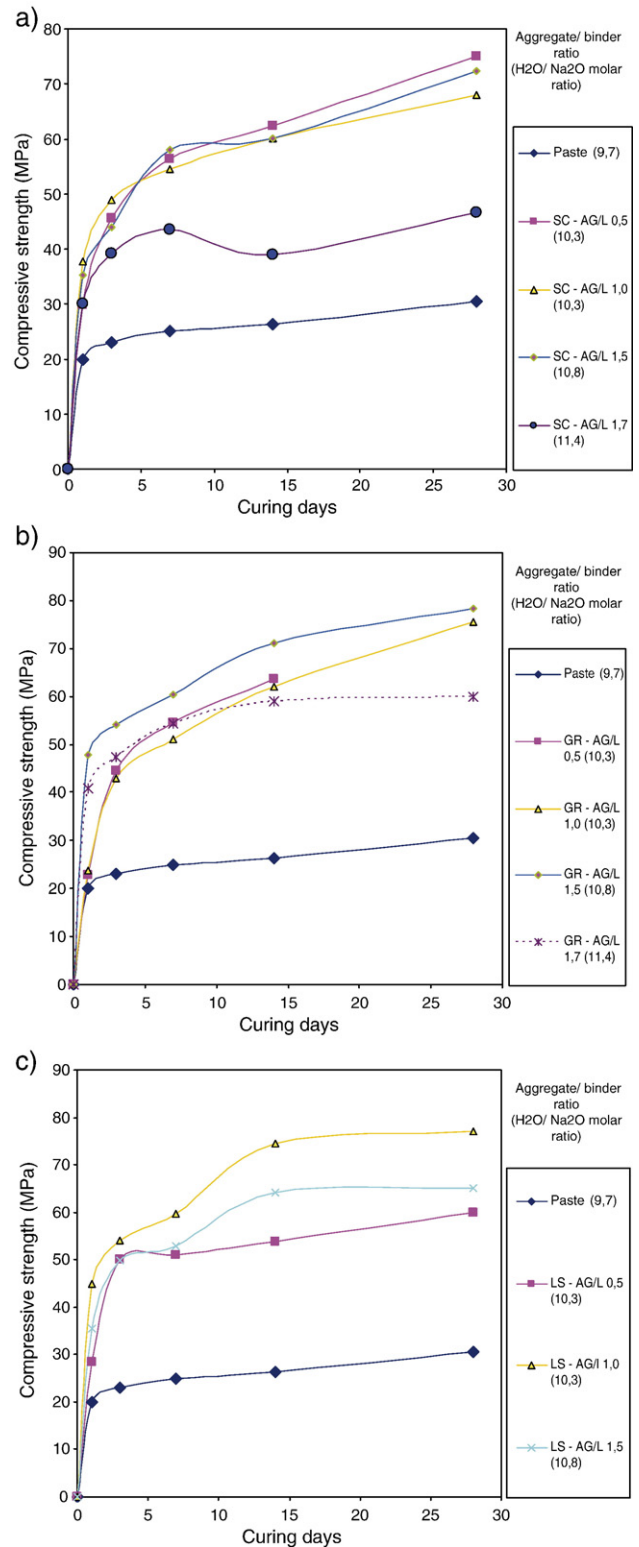| Samples	Binder		Activator		Aggregate
	90% Mine waste		28.57%NaOH	Extra water	
	10% Ca(OH)$_2$		71.43%Waterglass		
PCP	1		1	0.036	0
Ag/B=0.5	1		1	0.036	0.5
Ag/B=0.75	1		1	0.036	0.75
Ag/B=1.0	1		1	0.036	1.0
Ag/B=1.5	1		1	0.070	1.5

Fig. 1. Compressive strength according to aggregate/binder mass ratio and H$_2$O/Na$_2$O molar ratio in AAMWM mortars made with different aggregates; a) schist fine aggregates; b) limestone coarse aggregates and c) granitic coarse aggregates.

concrete however, Teixeira-Pinto [15] using alkali-activated metakaolin based mixtures found that some mixtures with calcium hydroxide and an atomic ratio of $SiO_2/Al_2O_3 = 5.1$ lead to higher compressive strength performances. However the final

SiO_2/Al_2O_3 atomic ratio in the hardened binder depends mainly on the reactivity of Al–Si because not all the silica and alumina are reactive so even despite the fact Al and Si have synchro-dissolution behaviour in alkaline solution, meaning they dissolve from the mineral in some linked form [2] one can not expect the same Si/Al ratio in the final hydration product as the one present in the original precursor material. Indeed most of the Al–Si materials cannot even supply sufficient Si in alkaline solution to start geopolymerization, this explains why they need extra silica provided in solution by waterglass, which influences the Si/Al ratio of the hardened binder.

Mine waste Blaine fineness is low but is in the range of the most used slag based alkaline binders. Despite the fact that increased fineness led to increase reactivity that can be overcome by using highly alkaline–silicate activators. Fernandez-Jimenez et al. analysed the joint effect of specific surface, curing temperature, nature and concentration of alkaline activator and found out that specific surface is the factor least statistically relevant, also reporting mechanical decrease with specific surface increase when using NaOH and waterglass activator [33].

3.2. Compressive strength

Compressive strength of alkali-activated mine waste mud (GMWM) binders is shown in Figs. 1 and 2. Plain geopolymeric mine waste mud paste was named PCP. GMWM binders using schist (Sc) fine aggregates with an aggregate/binder ratio of 0.5 were named SC-AG/B 0.5. Similarly when limestone (LS) or granite (GR) aggregates were used they were named respectively LS-AG/B 0.5 and GR-AG/B 0.5.

GMWM based binders showed very high strength at early ages, even higher than alkali-activated mixtures made with more reactive aluminosilicate raw materials like metakaolin [16]. It seems that strength level is due to calcium hydroxide and iron oxide which provides extra potential nucleation sites [34,35].

Compressive strength of GMWM binders made with schist sand are very similar when aggregate/binder ratio varies from 0.5 to 1.5 (Fig. 1). That assumption is not valid to an aggregate/binder ratio of 1.7 mostly because to keep a similar workability more water has been added to the mixture. Therefore when H_2O/Na_2O molar ratio is increased to 11.4 it leads to a strength decrease. Previous studies about geopolymeric mine waste mud mortars showed that H_2O/Na_2O molar ratio is a very significant parameter being closely related to compressive strength. Compressive strength increases due to less water and hence less porosity in the gel. And also when H_2O/Na_2O molar ratio decreases due to higher NaOH concentration [28]. Increasing NaOH concentration provides better dissolution of silicate and aluminate species leading to increase inter-molecular bond strength. The adverse effect reported by Lee et al. [34] related to reduction in strength due to excess of alkali has not been confirmed. This subject requires further investigations. However it must be a subject related to setting rate, because faster rates give less time for dissolution leading to more unreacted particles and strength reduction, so it may be expected that those authors used more reactive material (fly ash and kaolin) than mine waste mud used in the present work.

Similar behaviour happens when mixtures with coarse granitic aggregates are used, the only difference is that compressive strength of the mixtures with an aggregate/binder ratio of 1.7 shows a decrease in strength only after 14 days.
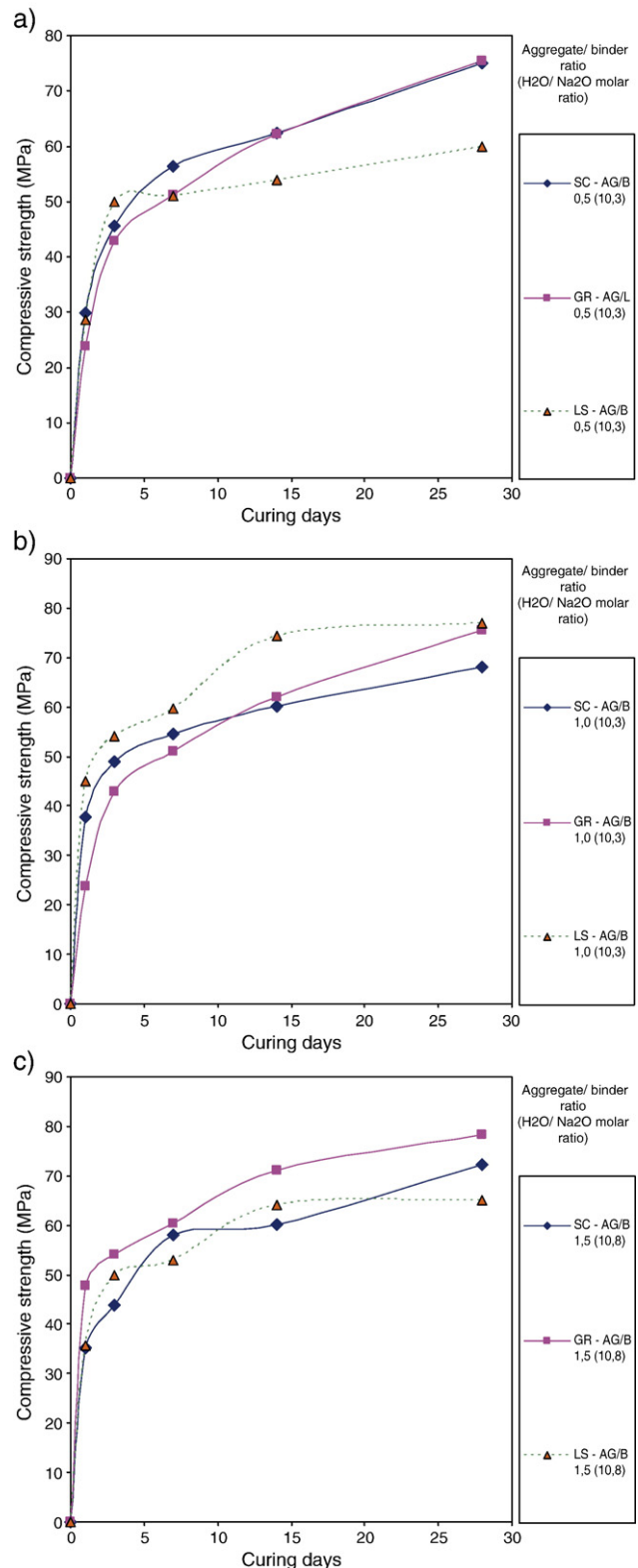

Fig. 2. Compressive strength in GMWM mortars made with different aggregate/binder mass ratio: a) 0.5; b) 1.0 and c) 1.5.
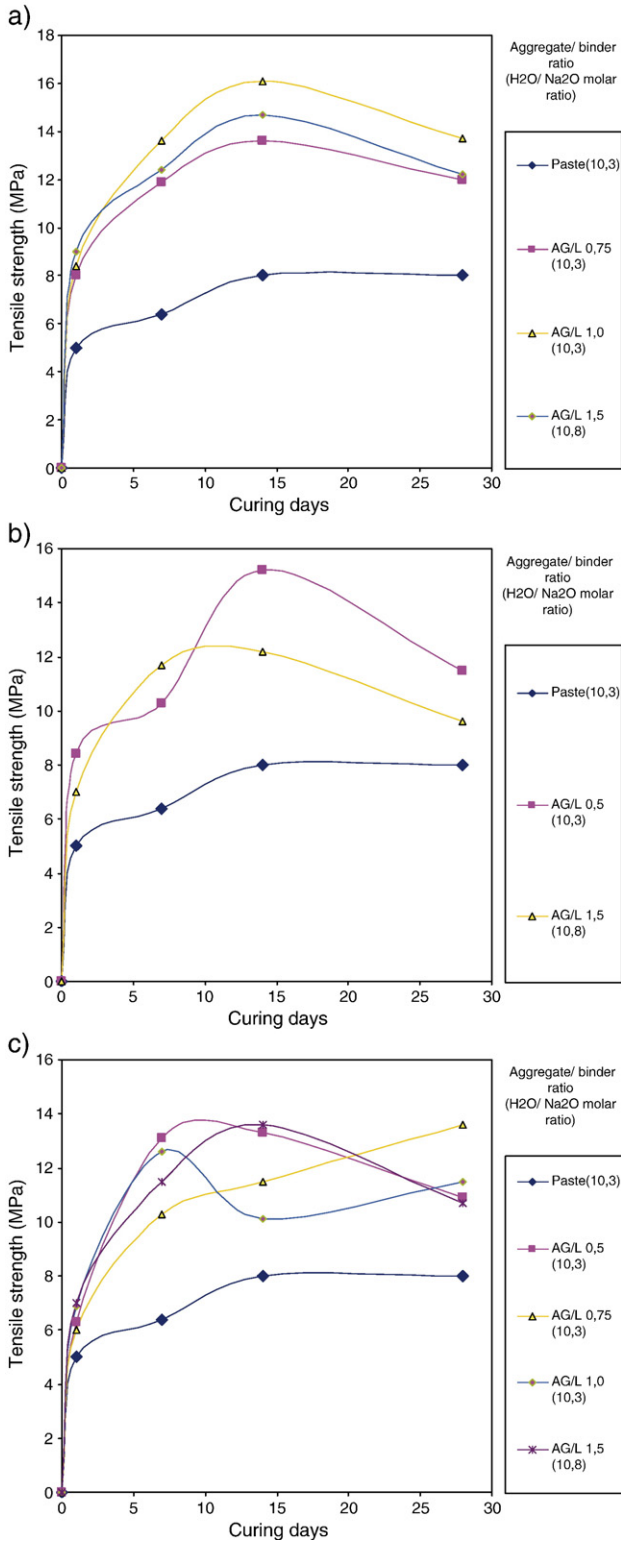
Fig. 3. Flexural tensile strength according to aggregate/binder mass ratio and H_2O/Na_2O molar ratio in GMWM binders made with different aggregates; a) schist fine aggregates; b) limestone coarse aggregates and c) granitic coarse aggregates.

Compressive strength of GMWM paste is significantly lower than compressive strength of GMWM mortars independent of aggregate type. In conventional concrete, the aggregates form a rigid skeleton of granular elements which are responsible for compressive strength. In the case of mortars with low content of aggregates i.e. aggregate/binder mass ratio below 2.0 aggregates do not form a rigid skeleton but a set of inclusions trapped in a continuous paste matrix. One suggests that strength behaviour can only be explained by the chemically active role played by the aggregates, which may be due to the dissolution of quartz in the presence of alkalis enhancing bond between paste and aggregates as reported by Shi et al. [23] even despite the low solubility of aluminosilicate minerals in alkaline medium. Furthermore the similar strength results obtained when using aggregates with different size and water absorption (schist and granite) prevents the explanation relying on the localized water near them, as it happens in OPC systems.

For an aggregate/binder ratio of 0.5 GMWM mortar mixtures with limestone aggregates achieve a maximum strength at early ages and that strength level keeps stable with curing age (Fig. 2). For the same aggregate/binder ratio silicon based aggregates (granite and schist) showed increased strength with curing time. That behaviour is somehow different from OPC systems where similar mechanical behaviour is presented by different aggregates [20]. In fact there's evidence confirming that aggregates have some influence in OPC binder systems as stated by other authors [21,22] but not at the same level presented in this work.

3.3. Tensile strength

Tensile strength of GMWM binders is shown in Fig. 3. Test results indicate that GMWM mortars possess very high tensile strength when compared to OPC systems (ft/fc ratio is around 10%). The bonding of GMWM binders to the aggregates is very high being confirmed by the fact that GMWM specimens submitted to flexural test, always show aggregates that have been cut and not pull-out as it happens in OPC binders. Tensile strength of GMWM paste is significantly lower than tensile strength of GMWM mortars being independent of aggregate type as seen previously for compressive strength, but different aggregates influence in a different manner tensile strength. For
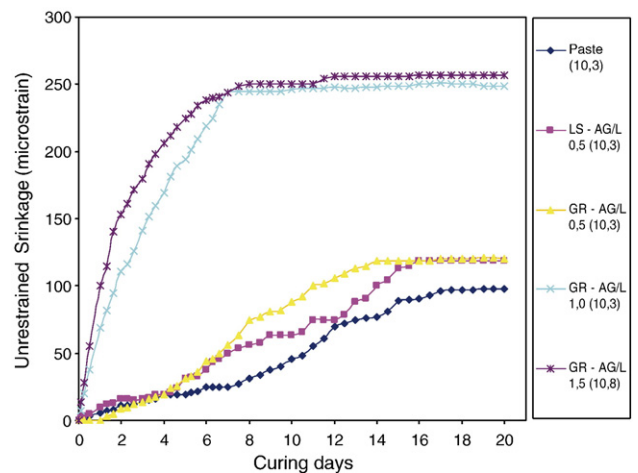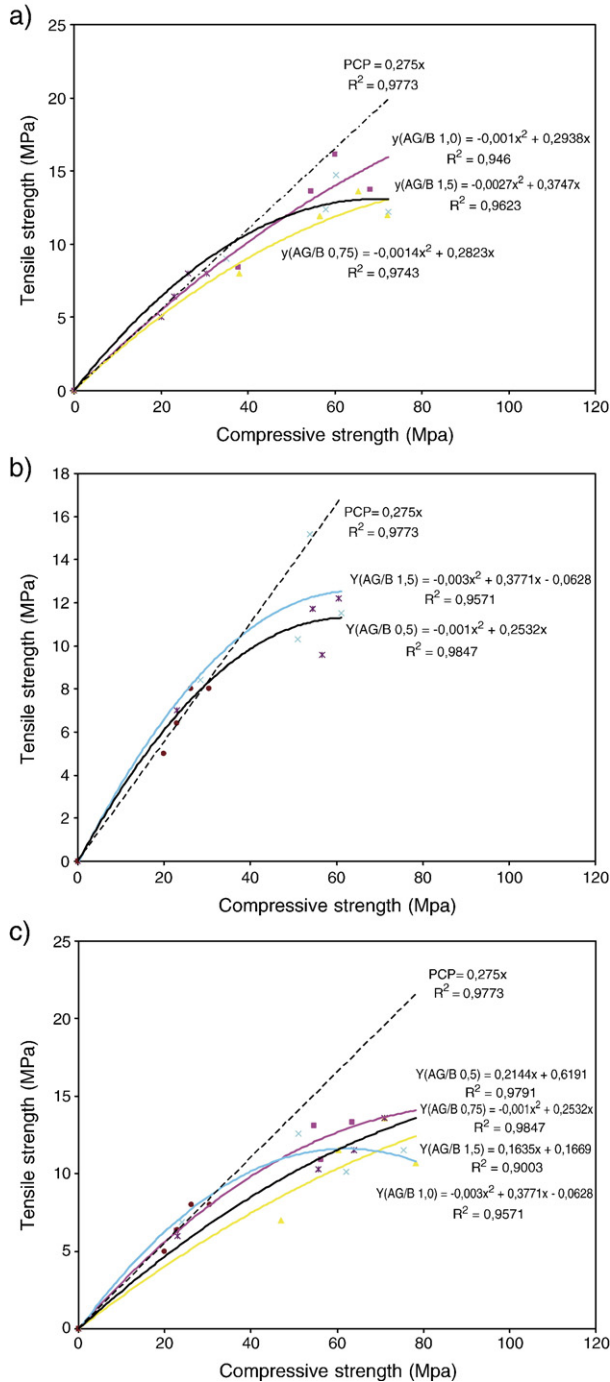

Fig. 4. Unrestrained shrinkage.

Fig. 5. Relation between flexural tensile strength and compressive strength according to aggregate/binder mass ratio and H_2O/Na_2O molar ratio in GMWM binders made with different aggregates; a) schist fine aggregates; b) limestone coarse aggregates and c) granitic coarse aggregates.

mixtures with fine schist aggregates an increase in aggregate content leads to an increase in tensile strength, however mixtures with an aggregate/binder ratio of 1.5 show a strength decrease due to an increase in H_2O/Na_2O molar ratio, from 10.3 to 10.8 when water had been added to the mixture to keep a similar workability. The same happens in mixtures with granite aggregates. However when using limestone aggregates the results are random with maximum tensile strength occurring at a

low content of aggregates and for 7 days curing, decreasing after that day, but the mixture with an aggregate/binder ratio of 0.75 present lower strength at 7 days curing, rising with curing time. The explanation may rely on the fact that strength evolution is somehow affected by shrinkage, which is currently associated with the presence of lime in alkaline activated binders. As it can be seen in Fig. 4 from shrinkage behaviour of mixes with granitic aggregates (GR-AG/L 0.5 and GR-AG/L 1.0) which has the same water content but a very different shrinkage level.

Tensile strength begins to decrease after 14 days of curing in all cases, except for mixtures with coarse granitic aggregates with an aggregate/binder ratio of 0.75, although it is not clear at that stage why that happens. Besides it must be noted that shrinkage shows a maximum at 14 days curing, so tensile strength decrease may be due to shrinkage cracks near the aggregates.
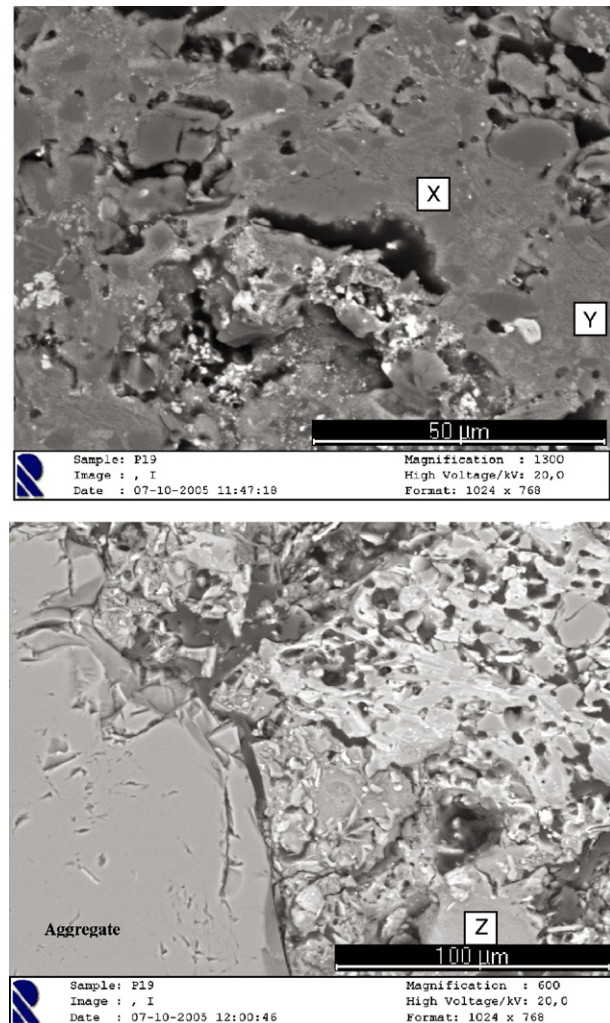

Fig. 6. SEM micrographs of GMWM mortar. The areas marked as F and G are identified as some type of aluminosilicate with the following composition: X[($CaO/SiO_2=0$); ($Al_2O_3/Na_2Oeq=2.5$) and $SiO_2/Al_2O_3=4$); Y[($CaO/SiO_2=0$); ($Al_2O_3/Na_2Oeq=3.1$) and $SiO_2/Al_2O_3=3.1$). The area marked as Z is identified as some form of calcium silicate with some sodium and with traces of aluminium in its composition ($CaO/SiO_2=0.8$); ($Al_2O_3/Na_2Oeq=0.3$) and $SiO_2/Al_2O_3=20.2$).
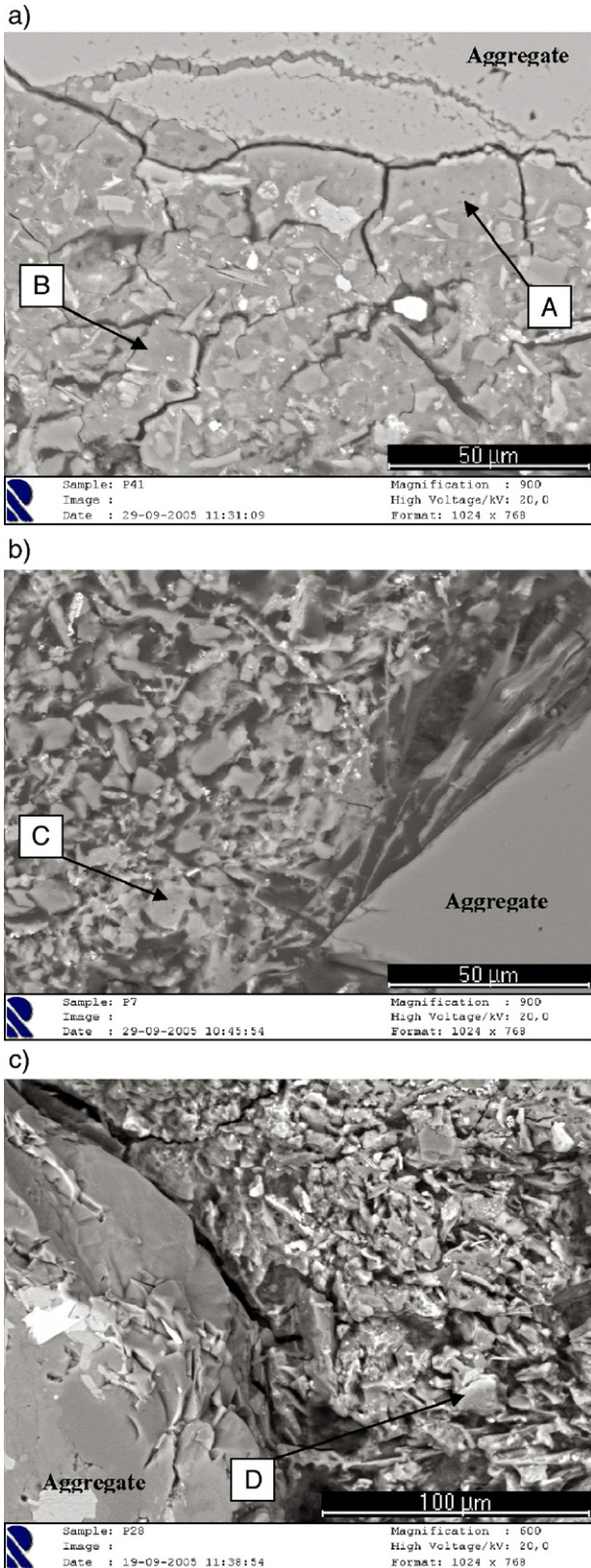
Fig. 7. SEM micrographs of interfacial transition zone in GMWM mortars made with different aggregates; a) limestone coarse aggregates; b) schist fine aggregates and c) granitic coarse aggregates.

GMWM mixtures with fine schist sand are the ones that present a regular behaviour, which is probably due to their lower size mixtures with a more regular distribution of aggregates throughout the matrix. Similar behaviour has been confirmed by Teixeira-Pinto when using geopolymeric metakaolin based binders [15]. That shows how much aggregate size influences tensile strength in GMWM binders, which is most likely due to the chemical interaction between the aggregate and the alkaline activated paste that has no comparison in OPC based systems.

Relations between flexural tensile strength (ft) and compressive strength (fc) of GMWM binders are shown in Fig. 5. Statistical correlations allow to conclude that aggregate content as well as aggregate type influences ft/fc ratio. It seems that strength results can't be explained by the differences in the fineness modulus of aggregates, instead it must be noticed that compositions with higher shrinkage display lower ft/fc ratio, which agrees well with the previous statement, about the influence of aggregates in tensile strength reduction.

3.4. Scanning electron microscopy analysis

Figs. 6 and 7 show SEM micrographs of GMWM binders. The analyses carried out by EDS on the different samples are displayed in Tables 5 and 6.

In Fig. 6 it is possible to see a scanning electron micrograph of a GMWM mortar sample, showing the existence of two different gel phases according to EDS analyses. This finding confirms the formation of geopolymeric gel (areas X and Y), as the main reaction product of Al–Si materials in presence of calcium hydroxide when submitted to high alkaline solutions, as stated by Alonso and Palomo [12]. High hydroxyl concentration prevents the calcium hydroxide dissolution, so only small amounts of calcium will participate in the formation of CSH.

The area identified as Z, has a low C/S ratio which means it as some sodium replacing Ca^{2+} in CSH; some authors have already demonstrated that sodium incorporation in CSH phase increased as C/S ratio decreases, and therefore they name it as NSH [36].

Aluminosilicates with similar SiO_2/Al_2O_3 and Al_2O_3/Na_2O molar ratios were found by Yip et al. [13,14]. Those authors also found the existence of an optimum of 20% (about 9% of calcium oxide) slag content in alkali-activation of metakaolin-

Table 5
EDS analysis

Oxides (%)	Mortar sample according to aggregate type			
	Limestone (LS)		Schist (SC)	Granite (GR)
	A	B	C	D
SiO_2	49.2	51.1	53.1	39.4
Al_2O_3	–	7.5	28.8	13.9
Fe_2O_3	–	4.8	3.5	27.4
K_2O	1.8	2.6	9.2	8.9
CaO	36.1	24.1	0.8	2.6
MgO	1.9	–	2.9	4.4
Na_2O	8.8	9.9	1.7	3.4
TiO_2	–	–	–	–
SO_3	2.2	–	–	–

Table 6
Atomic ratio analysis

Atomic ratio	Mine waste mud	Mortar sample according to aggregate type			
		Limestone (LS)		Schist (SC)	Granite (GR)
		A	B	C	D
SiO_2/Al_2O_3	5.5	–	11.6	3.3	4.8
Al_2O_3/Na_2Oeq	1.8	–	0.4	2.2	0.9
CaO/SiO_2	–	0.8	0.5	0.02	0.07
Na_2Oeq/CaO	–	0.25	0.4	8.8	3.1
Al_2O_3/CaO	–	–	0.2	19.6	3.0
MgO/Al_2O_3	0.2	–	–	0.25	0.8
Fe_2O_3/Al_2O_3	0.5	–	0.4	0.08	1.3

slag mixtures, leading to increase compressive strength, which according to them can be explained by the fact that formed CSH within the geopolymeric binder acts as microaggregates resulting in a dense and homogeneous binder. Furthermore increased calcium content leads to minor reactant aluminosilicate species dissolved. Be there as it may, is still not clear why calcium hydroxide plays such a significant role in the strength of geopolymeric binders. Fig. 6 shows SEM micrographs of the interfacial transition zone (ITZ) in mixtures of GMWM mortars made with different aggregates and Tables 5 and 6 show the results of SEM/EDS analysis.

In contrast with the porous typical interfacial transition zone of Portland cement mixtures, GMWM mortars present a very dense and uniform ITZ. The SEM images taken on the limestone based mixtures show that paste chemically bond to the aggregate. That explains why in limestone based mixtures, to the same H_2O/Na_2O molar ratio an increase in aggregate content from an aggregate/binder ratio of 0.5 to an aggregate/binder ratio of 1.0 leads to an increase in compressive strength. However clear cracks can be seen indicating shrinkage due to excess of calcium as suggested previously helping to explain some random strength behaviour.

The chemical composition of area A shows a silica gel with some sodium and some calcium, being some of the calcium due to the aggregate. As for area B, it seems to be made from silica gel with some sodium, aluminium and less calcium, meaning calcium content decreases with increased distance from aggregates. Area D presents a sodium content above the one presented by area C, that may be explained by the fact that aggregate distance is also higher and may be dissolution from aggregate could influence sodium incorporation.

The SiO_2/Al_2O_3 in areas marked as C and D is lower than in the waste mud, due to increased aluminium content. That behaviour is related to the silica and alumina reactivity of the original mineral as stated before; authors think that some aluminium will be dissolved from the aggregates to participate in the geopolymerization process, although further research is still needed in order to confirm that assumption. That hypothesis is consistent with the behaviour presented by binders with aluminosilicate aggregates, which present higher compressive strength. Other authors working with high SiO_2/Al_2O_3 molar ratio metakaolin also found out that hydration products

developed during geopolymerization have lower SiO_2/Al_2O_3 molar ratio than in the original precursor material [37]. Puertas et al. found similar Al/Si ratios in alkali-activated slag cement pastes and suggest that is due to substitution of Si by Al in tetrahedral positions in the silicate chain [38].

4. Conclusions

This study presents preliminary investigations about the effect of aggregates on the microstructure and mechanical behaviour of geopolymeric mine waste mud binders. Based on the experimental results the following conclusions can be drawn.

GMWM based binders showed very high strength even at early ages confirming the extreme reactivity of mine waste mud when activated with a highly alkaline solution. The addition of aggregates increases strength behaviour acting almost as a catalyst. It's suggested that may be due to the dissolution of quartz and alumina in the presence of alkalis enhancing bonding between paste and aggregates. Other hypotheses related to aggregate type could not be confirmed.

Unlike OPC systems GMWM based binders possess extremely high tensile strength. Tensile strength begins to decrease after 14 days of curing although it is not clear why that happens. It is suggested that may be due to shrinkage cracks near the aggregates. In alkali-activated based binders schist and granitic aggregates are chemically reactive increasing strength with curing time. A dense and uniform ITZ was detected in GMWM based binders. Mixtures with limestone aggregates showed that GMWM paste chemically bond to the aggregates but present higher shrinkage which may explain random strength behaviour.

In order to confirm the results obtained in the present study further research is still required using aggregates with different sizes and different water absorption.

Acknowledgement

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