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RECYCLING OF HIGH QUANTITIES OF WASTEPAPER SLUDGE ASH FOR PRODUCTION OF BLENDED CEMENTS AND ALTERNATIVE MATERIALS

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Abstract

Among low cost and widely available raw materials, wastepaper sludge ash is of key interest as potential construction material. These materials come from the paper recycling industry. In present study, wastepaper sludge ash (WSA) retrieved from UPM Chapelle Darblay Company located in the south of Rouen, France, were subjected to preliminary physicochemical characterization by X-Ray fluorescence, laser granulometry and scanning electron microscope (SEM) imagery for particles morphology. Afterwards, the WSA considered as mineral addition was blended with cement, lime, and calcined kaolin for the highest possible dosages in WSA. The effects of reactivity were evaluated by means of mechanical strength measurements. Cylindrical samples were prepared with WSA mixtures and air-cured for 7, 14 and 28 days at ambient room temperature. WSA content ranged from 30-95%. The samples obtained were subjected to unconfined compressive strength to verify the occurrence of reactivity and resistance. The results indicated that the strength increased along the curing time for all mixtures except for the reference sample in which 100% of WSA was used. The lime addition does not impact the unconfined compressive strength. As main results, on one hand, WSA and cement based mixtures with a weight ratio of 95/5 develop 2 MPa compressive strength and more during the curing time which suit for construction materials. On the other hand, with only 5% of calcined kaolin, a value of 9 MPa was attained after 28 days indicating that mixtures with calcined kaolin can be effectively used for building materials such as bricks or blocks.

Keywords:

Wastepaper sludge ash, lime, cement, calcined kaolin, unconfined compressive strength, morphology.

Highlights

1. Possible implementation of high volume of wastepaper sludge ash (WSA) in mixtures blended with cement and/or lime, or with calcined kaolin.
2. Wastepaper sludge ash and cement based mixtures with a weight ratio of 95/5 developed an unconfined compressive strength (UCS) more than 2 MPa after 28 days and are suitable for construction materials.
3. Wastepaper sludge ash and calcined kaolin based mixtures can also be employed for building materials. Their UCS strength was nearly 9 MPa after 28 days when only 5% of calcined kaolin was added.
4. Effect of UCS strength is related with density of samples, i.e. with compaction, thus, suggesting that compaction should be considered for manufacturing of materials because it contributes to improve the strength.

1 INTRODUCTION

Wastepaper sludge ash (WSA) is a residue which results from co-incineration of paper sludge generated by the paper mill, recycling paper industry and waste of wood biomass. Thousands of tons of paper sludge are produced every year around the globe and as a result, thousands of tons of WSA ash. In Europe, pulp and paper industry generate around 11 million tons of waste annually, 70% of which is derived from the production of deinked recycled paper [1]. According to the Confederation of European Paper Industries (CEPI) and International Council of Forest and Paper Associations (ICFPA), the paper recycling rate in Europe reached approximately 72.3% in 2017 [2]. UK produces over 1.5 million tons of wastepaper sludge annually for 125,000 tons/year of WSA [3-4]. In France, the WSA production for the United Paper Mills Ltd (UPM) and Norske Skog Goble (NSG) recycling paper industries is around 60,000 tons (reduced by the half since 2017) and 50,000 tons every year, respectively [5-6].

Large volumes of ashes are still difficult to recover even if potential recycling routes for wastepaper sludge ash (WSA) have been investigated such as soil amendments, soils stabilization, road sublayers, partial replacement of cement in concrete industry, manufacture of plaster blends of blocks and lightweight aggregates [7-13].

Sometimes paper sludge is used after specific treatment as an absorbent material such as the absorption of pollutants in the case of ammonium (NH_4^+) present in municipal, industrial, and agricultural wastewaters and leachate [14]. Till now a part of WSA continues to be disposed-off or landfilled.

The reuse of WSA in an economical and environmentally sustainable manner for manufacturing alternative materials is always a challenge. The environmental quality of paper sludge and WSA have been investigated by Boni et al., [15] and Zmamou [16] respectively, demonstrating the non-hazardous nature for the environment in terms of heavy metal release. Based on these environmental considerations and due to the chemical properties of the fly ash, WSA is considered as an alternative source of sustainable waste material in the construction industry. In addition, it is estimated that the annual production of OPC will increase by approximately 50%, starting from the year 2017 to the end of 2050. Meanwhile, the manufacturing of 1 ton of OPC is estimated to release approximately 1 ton of carbon dioxide (CO_2) [17]. So, using of other alternative materials like WSA constitutes an environmental and economic solution.

Most research work has shown that there are two ways of recovering WSAs, either by adding them to Ordinary Portland Cement (OPC) or by partially substituting cement in mortars or concrete [18-36]. In particular, WSAs are used in mortars for masonry to support green technology as carbon dioxide emissions can be reduced since lower cement production is involved [37]. Concerning the recent above-mentioned studies, a maximum amount of WSA is reused ranging from 50% to 100%, with very high cement addition levels till 50%. Usually a microstructure analysis is also coupled with environmental and mechanical properties during the implementation of such studies.

Nevertheless, the main technical obstacles to the WSAs recovery are the presence of free lime which by excess causes problems of swelling and their activation. Very few studies focus on the activation of WSAs [38] even though this technique is widely used for coal fly ash and in the development of geopolymers [39]. In the present study the choice not using the activators usual for fly ash *i.e.* NaOH , KOH , and Na_2SiO_3 , is to reduce the cost of the WSA-based recovered product, to avoid handling chemicals and to use first traditional and commercial binders such as cement, lime and calcined kaolin. This last additive makes it possible to increase the compressive strength. Usually this also possible by incorporation of a coarser grain size corrector as a sand and if possible, a waste sand to preserve natural resources [40-41].

As observed by de la Villa et al., [42], the WSA obtained after incineration is combined with reactive silica, alumina and lime, where the chemical composition remains similar as that of Portland cement components. Bai et al., [43] observed that the presence of CaO in WSA has both negative and positive effects for the use of WSA as a hydraulic binder for the hydration post-setting.

However, for practical applications, WSA ash needs further investigation to become attractive technically, economically and eco-friendly and to be rendered as more market appealing. Following the European directive [44], the current study focuses on elimination of WSA waste and recycling at a possible large scale. Depending on its physical and chemical properties, WSA is considered as a hydraulic binder in different mixtures in which WSA is the major component, whilst additives are added to control the hydration post setting effect [43] and to enhance compressive strength.

The present study was designed to reduce the amount of WSA waste in the paper industry by investigating new recycling routes and also reducing the use of natural resources. The main objective was to evaluate the feasibility and the implementation of a high volume of wastepaper sludge ash in mixtures blended with cement and/or lime, or with calcined kaolin. Then, the additives influence on WSA based mixtures *i.e.* *cement and calcined kaolin*, has been investigated through the unconfined compressive strength (UCS), the density and the morphology. Considering their mechanical properties, possible WSA recycling routes could be proposed as for addition of cement with a weight ratio of 95/5 suitable in nonstructural materials and for the addition of only 5% of calcined kaolin enhancing the UCS, construction materials may be considered.

2 MATERIALS AND EXPERIMENTAL TESTING

2.1 Raw materials

The WSA was provided directly by the French company UPM Chappelle Darblay, located in Grand-Couronne, near Rouen harbor (France). To burn wastepaper and other inputs, the fluidized bed technology was applied as recommended by Directive 2000/76/EC [45]. So, the sludge remains in the combustion zone for a few seconds at 850 °C, the flue gas is filtered and the WSA is recovered. A quantity of 60 000 tons/year of WSA is produced and stored in silos.

The WSA recycling consists of mixtures of WSA and mineral additives without any treatment. WSA based mixtures are composed either with cement (C) or with calcined kaolin (CK). In both mixtures, lime had also been added. Some reference mixtures with only WSA as a component have been made to investigate WSA binder ability and WSA efficiency in different mixtures tested. WSA dry material was provided by UPM paper mill (Rouen, France) which uses 100% recycled paper as the raw material [5]. WSAs were obtained from the combustion process of de-inking sludge with wood waste. They are classified as F class for fly ash according to ASTM [46] and as non-hazardous waste [44].

All WSA-based mixtures proposed are composed with:

- Wastepaper Sludge Ash (WSA), main component of mixture recovered as a fine powder.
- Ordinary Portland Cement (OPC), referenced as a CEM II B-M (S-LL) 32.5R, with a mean diameter D_{50} of 507 μ m. As this study is part of a sustainable development context, so the type of cement employed was of commercial origin and supplied by a local cement plant in the same region where the wastepaper sludge ash was acquired. Although, a CEMI type cement would have been preferable as cement reference, but the cement plant had only a CEMII type of cement which already contains mineral additions other than clinker.
- Commercial quicklime L, graded as 0/1.5 mm lime with a CaO percentage exceeding 93%, and a mean diameter D_{50} of 567 μ m.
- Calcined Kaolin (CK), a commercial fine powder with a mean diameter D_{50} of 50 μ m,
- Tap water.

After selecting all the constituents, a complete characterization of the mixture was carried out for its chemical, physical, and morphological properties. The wastepaper sludge ash as main component in mixtures investigated is more detailed and only partial characterization is reported for mixtures additives *i.e.* cement (C), lime (L), calcined kaolin (CK).

2.2 Specimen preparation and mixture

The sample preparation consists of 3 steps:

- Mixture preparation using a laboratory mixer: Its first involved weighing various constituents of the mixture and pouring them into the mixer. Afterwards, these were dry homogenized. Water was added carefully in order to control the consistency of the mixture.
- Compaction and molding of samples: In previous studies, three methods of compaction for fine soils mixed with binders have been tested and compared by Liu [47]. These methods consisted of coring at the desired sample shape, dynamical compaction as the standard Proctor procedure, manual compaction and picketing. The last method was developed by Duan [48], considering the following points (i) very near compressive strength obtained for samples of same mixture, (ii) easier to prepare and repeatable, (iii) time saving for preparing a specimen. It has been concluded that this is the best method. This method of compaction was selected as it was also assumed to be more representative for implementing on road layers so was employed in present study.
- Air curing period till testing at 7, 14 or 28 days: After demolding, the samples were stored and cured at laboratory temperature, 20 °C \pm 2 °C.

The specimens used are cylindrical and have a slenderness ratio of 2, in accordance with the standard NF P 98-230-2, 1993 [49]. The dimensions of mold specimens are 34*68 mm (diameter* height). Three samples are tested as required by standards for any mixture.

As said above, in the present study, two hydraulic mineral matrices were investigated: (i) WSA-Cement+/-Lime and (ii) WSA-Calcined Kaolin+/-Lime. The cement or lime additions in the first series of mixes were 0, 5 or 10 wt%. The different mixes and their references are presented in table 1. For the second series of mixes, calcined kaolin varied from 5 to 40wt% and a constant of 30wt% of lime was added. Compositions and references are shown in table 2. Ratios of Water/Binder are also indicated in tables 1 and 2.

2.3 Experimental testing

The ratio Water/Binder (W/B) of each mixture was determined in order to keep constant workability of mixtures. The unconfined compressive strength (UCS) tests were performed on cylindrical samples for all mixtures at different dates *i.e.* 7, 14, 28 days in air curing conditions at 20 °C \pm 2 °C.

Mixtures	WSA ¹	C ²	L ³	W/B ⁴
WSA	100	0	0	0.5
WSA-C5	95	5	0	0.6
WSA-C10	90	10	0	0.6
WSA-C5-L5	90	5	5	0.6
WSA-C5-L10	85	5	10	0.6

¹Wastepaper Sludge Ash, ²Cement, ³Lime, ⁴Water/Binder

Table 1: WSA based mixtures with cement.

Mixtures	WSA	KC	Lime	W/B
WSA	100	0	0	0.5
WSA-CK40	30	40	30	0.7
WSA-CK20	50	20	30	0.7
WSA-CK10	60	10	30	0.6
WSA-CK5	65	5	30	0.4

Table 2: WSA based mixtures with calcined kaolin.

The UCS is the main mechanical parameter in the experimental program from which two types of WSA based mixtures must be analyzed and compared.

3 WSA CHARACTERIZATION

3.1 Chemical composition

The chemical composition of WSA powder was determined by X-Ray fluorescence (XRF) analysis. X-ray fluorescence spectroscopy is a non-destructive method. The WSA powder was compacted to obtain pellets. Mapping of a surface of WSA-pellet has been performed to obtain the chemical elements present in WSA. The source used is Rhodium (Rh) with X-ray beam size of 30 μm . Conversion factor is used for various elements to obtain oxides content. The oxide composition of the wastepaper sludge ash used for the mixtures is shown in figure 1.

Wastepaper sludge ash is mainly composed of calcium oxide (CaO), silica (SiO₂) and alumina (Al₂O₃). Other constituents are considered as minor ones (MgO, SO₃, K₂O, TiO₂, MnO, P₂O₅). According to the presence of oxides and their amount, WSA can be compared to the cement (hydraulic binder) or to the fly ash (pozzolanic binder). This comparison allows investigation of the potential reuse of WSA as a binder [50].

WSA is very rich in calcium oxide (>50%). According to the chemical composition obtained, the nature of fly ash is silica-calcareous type. The total percentage of three major oxides (CaO, SiO₂, Al₂O₃) accounts for nearly about 92% as shown in figure 1. The calcination of wastepaper sludge ash (WSA) at 480 °C during 6 hours provides a very low organic matter content (1.4%), that corresponds to the UPM Company commercial reference product given as *Calcichap* as inorganic fly ash [16].

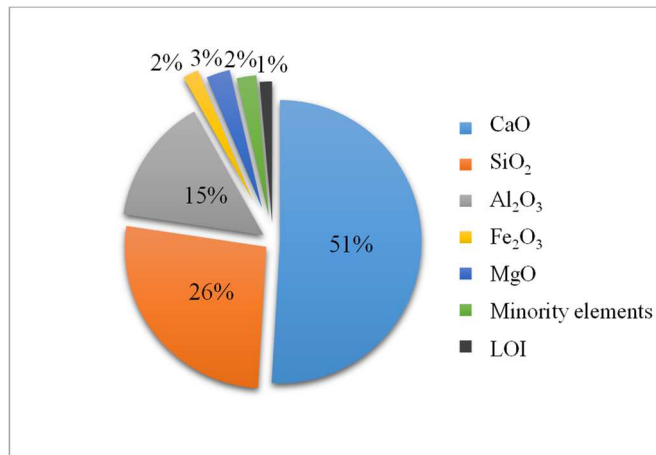


Figure 1: Chemical composition of WSA.

The chemical composition of WSA produced by UPM Chappelle Darblay in 2013, was measured and compared with different chemical compositions of WSA available in literature, as shown in figure 2. All the results of chemical composition of WSA used were obtained from the following references [13, 28-30, 43, 51-59]. In figure 2, we have chosen to write the main author's name with the year of publication in order to show the variability of material according to the source and year of investigation. It is clearly observed that the composition of major oxides varies from 77% to 92%, showing overall consistency of the WSA composition in the world.

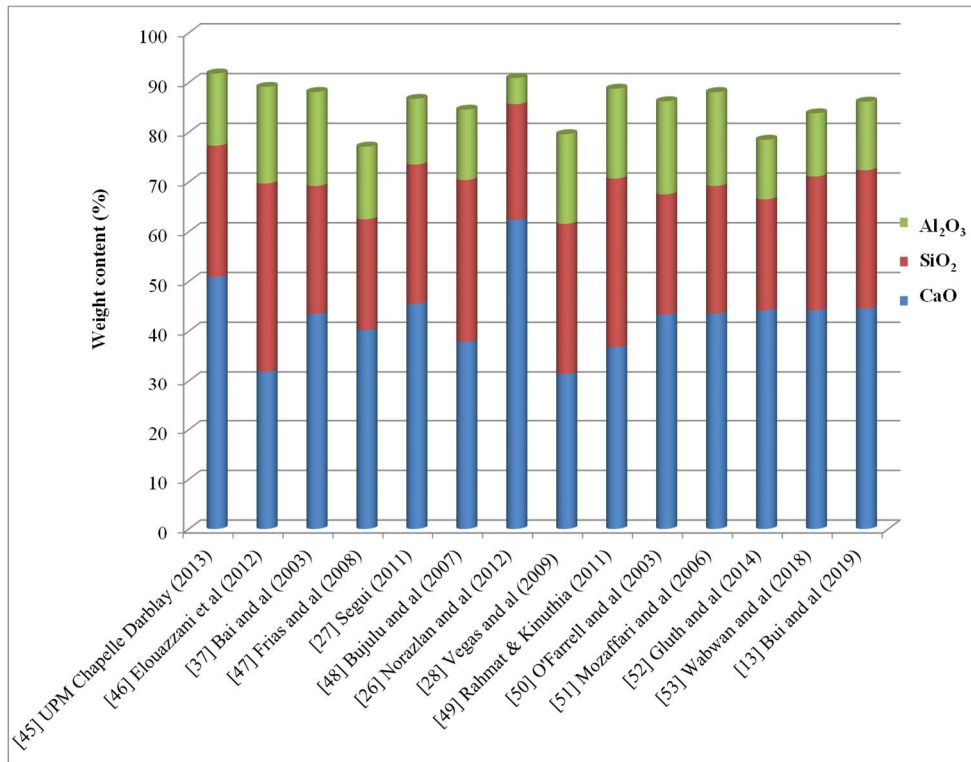


Figure 2: Chemical composition of different WSA investigated around the world.

3.2 WSA grain size distribution and morphology

In accordance with the environmental laws, French recycled paper industries must continuously control WSA characteristics along their production (chemical composition, grain size, etc.). When a low variation in these parameters is observed, it is considered as negligible. Figure 3 demonstrates the grain size distribution obtained by two laboratories (Lab U *i.e.* Unilasalle, April 2013 and Lab M. *i.e.* M2C, May 2013). The two-particle size distributions were determined by laser granulometry at two different dates. Figure 3 clearly shows that WSA distributions are similar with a mean diameter of wastepaper sludge ash estimated to be 290 μm . About 80% of ash has a fine particle size below 600 μm . Furthermore, a consistent WSA production is confirmed. This consistency is considered very important in a future industrial process for manufacturing WSA-based materials. Some WSA grain size properties obtained according to the French standards are given in table 3. But physical characterization includes morphology, ignition loss, actual density, Blaine specific surface area, and particle morphology analysis (granulo-morphology). These parameters were chosen because they have a significant impact on the reactivity of WSA in cementitious matrices. These parameters they play a role water demand, so in the workability of matrices. To summarize the main physical properties of WSA are listed in table 3.

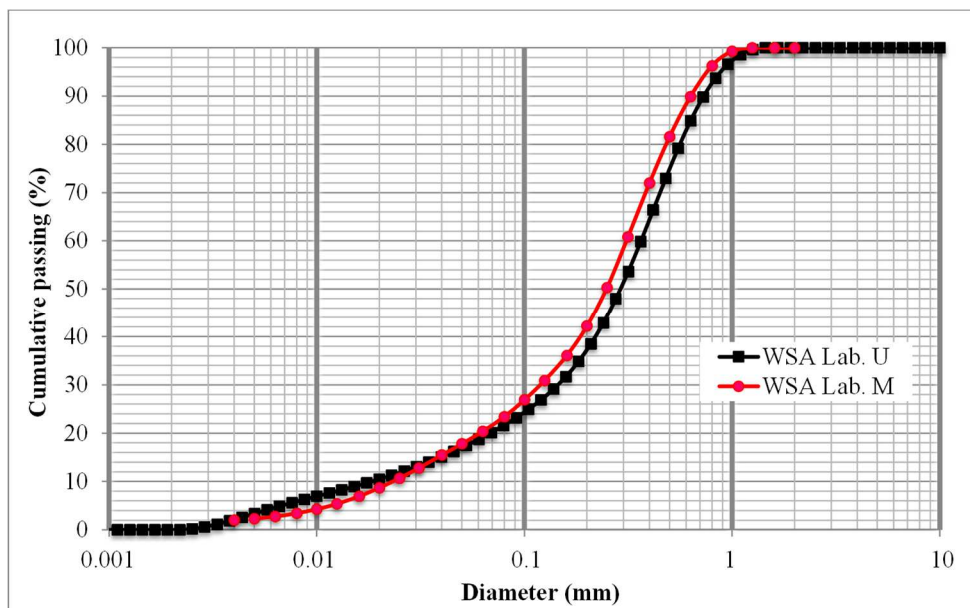


Figure 3: Typical WSA grain size distributions.

Parameters	
Solid particles density (g/cm ³)	2.87 [38]
BET surface area (m ² /g)	2.12 [38]
Bulk density (t/m ³)	0.923
Particle size D ₅₀ (μm)	290 μm
Coarse grain (>2000 μm) (%)	0
Medium grain (80-2000 μm) %	78
Fine grain (2-80 μm) %	21
Very fine grain (<2 μm) %	1
Relative humidity HR (%)	0.1
LOI (%)	1.4
Color	Grey
Oder	Oderless

Table 3: Main physical properties of wastepaper sludge ash.

The specific surface of WSA was evaluated by the BET (Brunauer-Emmett- Teller) method [38]. WSA has a significantly higher specific surface, which should imply a very high water demand and, this has been observed during the preparation of mixtures.

WSA bulk density is less than 1 t.m⁻³ compared to constituent values for mixtures used as binders (2100-3100 kg.m⁻³). Monosi and *al.* [18], observed that the bulk density of the paper ash is equal to 1200 kg.m⁻³ which is lighter than ordinary sand (that predictably ranges from 2500 to 2600 kg.m⁻³) and binders (2100-3100 kg.m⁻³). The solid particles density of WSA, measured with a helium pycnometer, shows that the value varies between 2500 and 2900 kg.m⁻³ as determined by Davidenko, [60]. The bulk density of the WSA mixed is 0,923 t.m⁻³ where, the relative humidity after 24 hours in the oven at 105 °C was 0.1%. All WSA was dried before mixing samples for the 2 test series.

WSA grain morphology observed by scanning electron microscope (SEM) images is illustrated in figure 4. WSA exhibited grains of irregular shapes with opened or closed spheres. This explains the high capacity of water absorption of WSA.

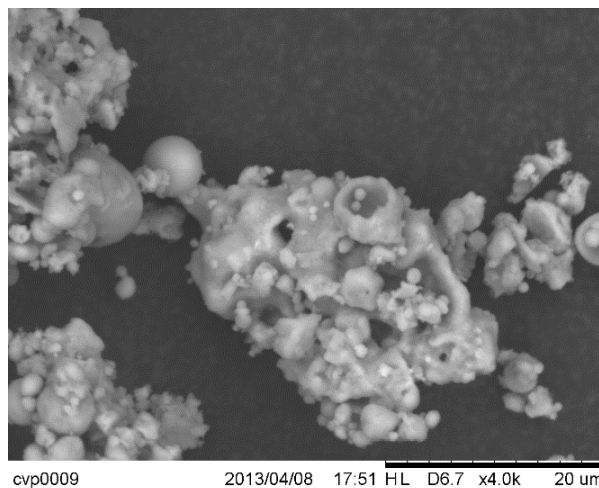


Figure 4: WSA grain morphology by SEM images.

3.3 Comparative grain size distribution of mixture components

Figure 5 presents the grain size distribution (GSD) of mixed components, also determined by laser granulometry.

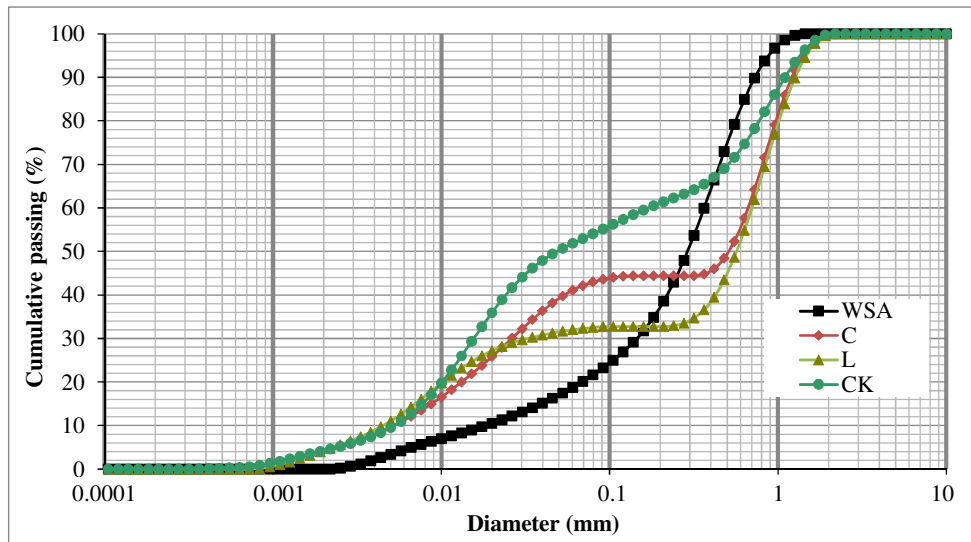


Figure 5: Components grain size distributions in mixtures.

The grain size of all additions is less than 2 mm, so all mixtures have a fine granulometry while, cement and lime powders are not well graded.

4 MECHANICAL STRENGTH

4.1 Unconfined compressive strength (UCS)

UCS maximal average values obtained for 3 samples at different ages for both WSA Cement-based mixtures and WSA-Calcined Kaolin mixtures are illustrated below in figures 6 and 7, respectively. The water demand depends upon mixtures considered as indicated by the Water/Binder (W/B) ratio values. UCS resistance ranges from 1 to 9 MPa considering all mixtures.

For WSA-C-L mixtures, UCS value after 28 days was approximately 2.5 MPa with only 5% and 10% of cement. But UCS tends to drop below 2.5 MPa with the addition of lime. It can be partly explained by the composition of WSA. WSA powder contains more than 50% of CaO, part of which is free lime. So, addition of lime is not necessary, WSA content 3.8% of free lime.

Concerning the UCS results of WSA-CK mixtures, UCS resistance relies on the percentage of calcined kaolin being added. Only 5% of CK gives the highest UCS value. Beyond 5%, the UCS falls down and reaches a value of 2 MPa whatever the addition level from 10 to 40% addition. This drop is observed along the curing time. An optimal CK amount value is detected around the 5%.

For mixtures with calcined kaolin, it is not possible to keep the W/B ratio constant for all mixes, because CK additive has a higher capacity of water absorption. For example, for WSA-5CK mix with a W/B ratio of 0.4, the mixture is low workable. To improve the workability *i.e.* water demand, W/B increases but when W/B ratio is greater than > 0.4 , the mixture tends to be quickly wetter. When CK level increases, water demand is adjusted as shown in figure 7 and consistency was kept constant.

Cost of calcined kaolin is almost twice as compared to that of cement. Regarding intended application and considering the mechanical resistance (UCS) and the cost of various additives, the mixtures with 5% of cement or calcined kaolin are sufficient for proposing a WSA-based material for a road sublayer or construction material, respectively. Recall that a value of 1 MPa could satisfy requirements for road sublayers.

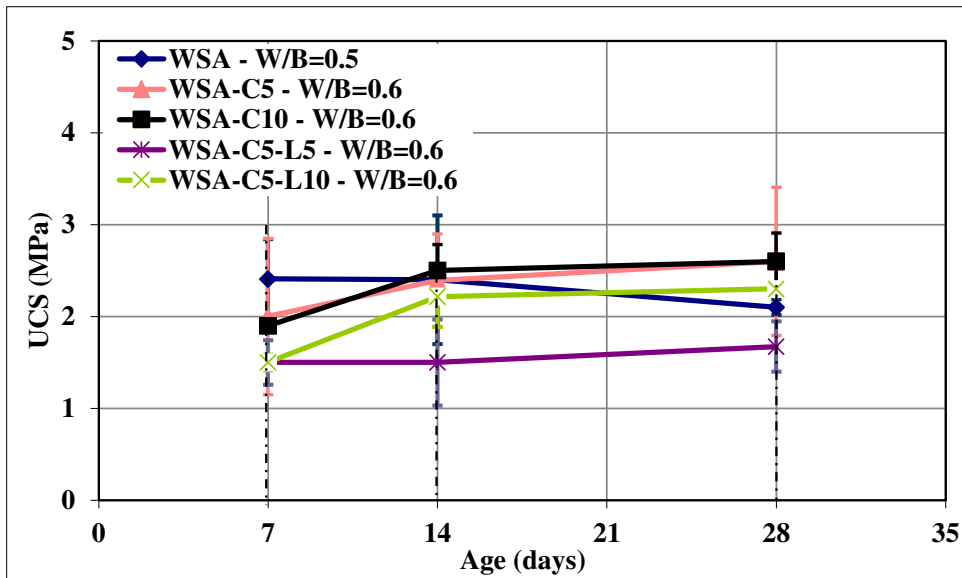


Figure 6: Cement and lime addition effect on UCS of WSA-based mixtures vs time.

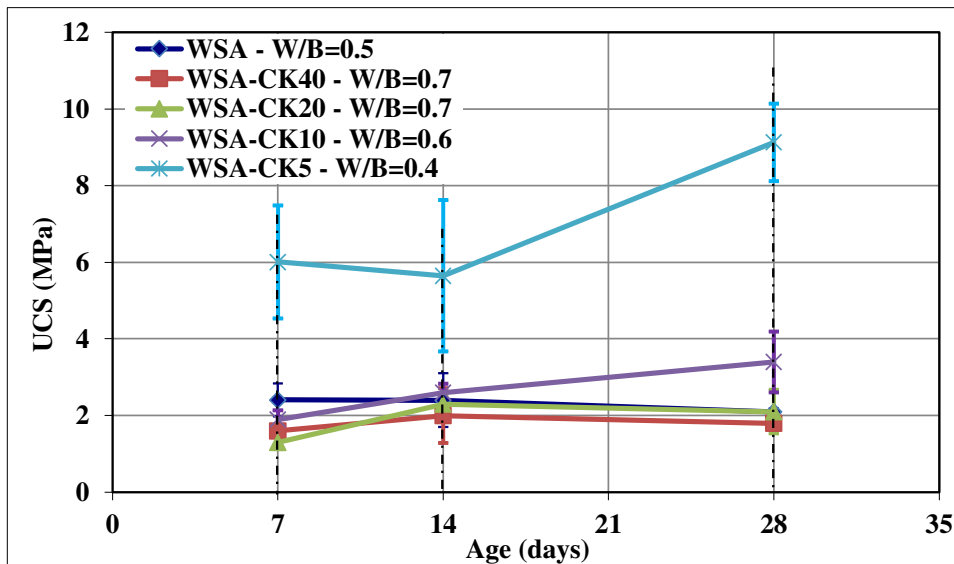


Figure 7: Calcined kaolin addition effect on UCS of WSA-based mixtures vs time.

Mixtures (WSA-C5 and WSA-C10) with 5% and 10% of cement respectively and W/B=0.6 have the same trend, thus achieving the similar UCS value. In order to use a minimal quantity of additives, the mixture with only 5% was chosen. These results indicate that the mechanical strength depends on the quantity of water added and the age of samples. UCS tends to increase over 28 days for the two mixtures with 5% of cement or calcined kaolin. Figure 8 shows the effect of lime and calcined kaolin addition on mechanical strength at 28 days. As observed, lime does not affect the strength considerably, unlike calcined kaolin.

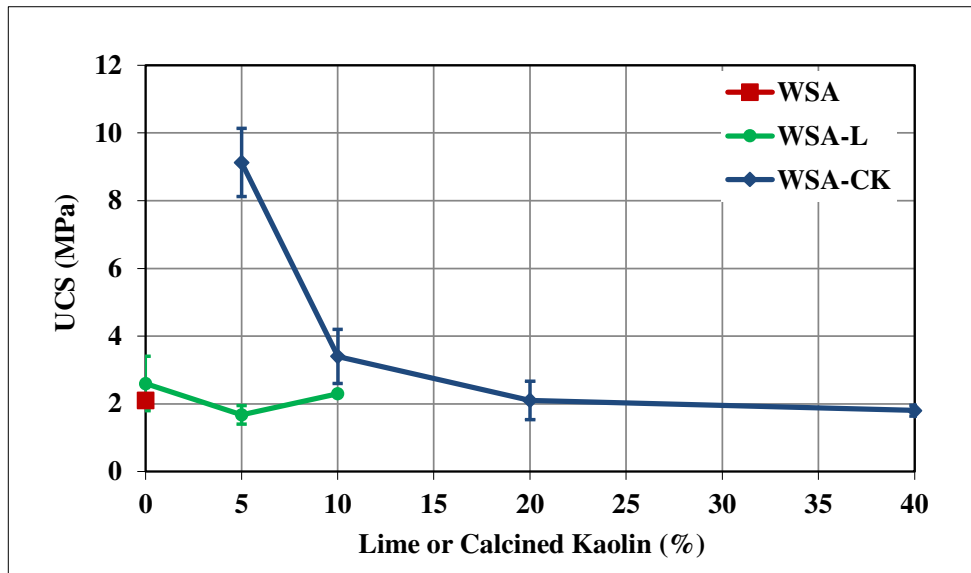


Figure 8: Effect of lime or calcined kaolin addition on UCS of WSA-based mixtures at 28 days.

4.2 UCS-bulk density relationship

Bulk density was determined from mass and dimensions of samples before testing. UCS at different days of curing *i.e.* UCS for 7, 14 and 28 days is related to bulk density of material in figure 9: a) Mixtures WSA-Cement-Lime, b) WSA-CK-Lime. On the one hand, density according to every added component varies and depends on the hardening of the mixture. So UCS relies on the density of materials. We obtained two UCS ranges with the two series of mixtures being considered. The values for WSA-C-L mixtures matrix ranged from 1.5 to 3 MPa, while reached up to 9 MPa in case of WSA-CK mixtures.

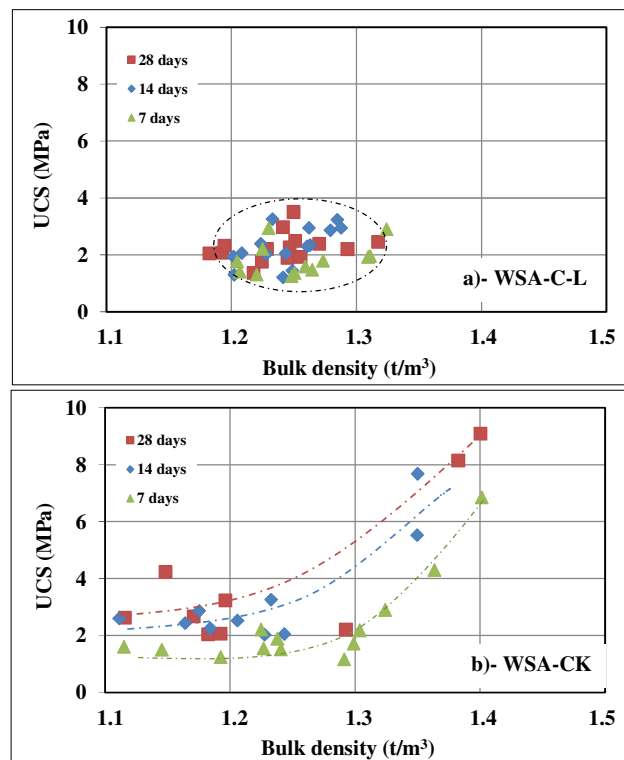


Figure 9: UCS- bulk density relation for WSA-based mixtures: a)- WSA-C-L, b)-WSA-CK

4.3 UCS-morphology consideration

In this section the morphology of the two mixtures with highest strength *i.e.* WSA-C5 and WSA-CK5, was analyzed in order to explain the difference of UCS resistance values obtained. SEM image technique was used as shown in figure 10. Mixtures were observed at curing ages of 7 and 28 days.

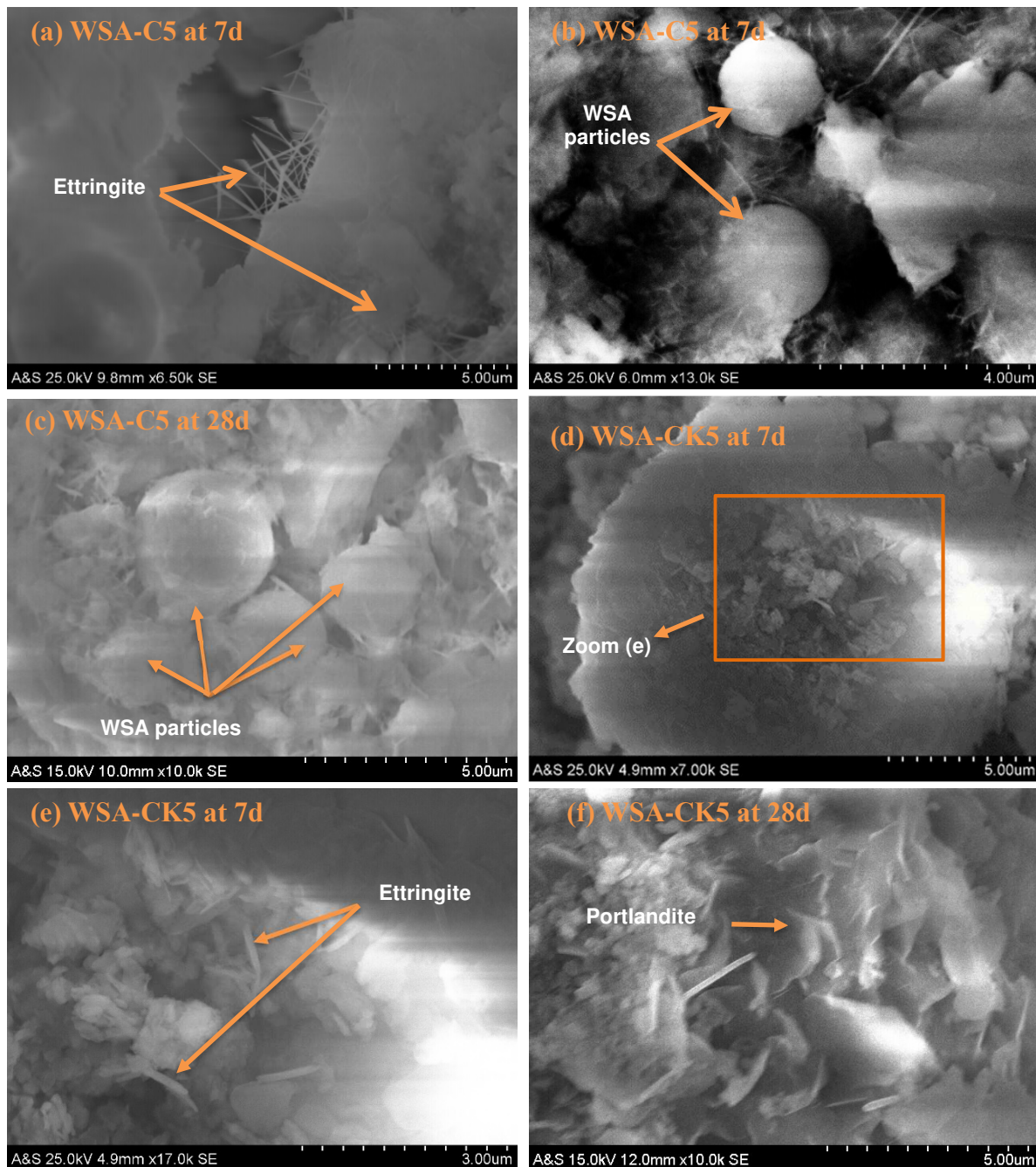


Figure 10: Morphological structure of WSA-C5 and WSA-CK5 at 7 and 28 days (day=d on figures) of curing by SEM.

Observations revealed the presence of various mineral phases such as portlandite and ettringite in several forms (free or oxide). SEM images clearly depict the presence of WSA spherical grains concerning the WSA-C5 mixture. It indicates the presence of non-hydrated grains of WSA in this mixture. Consequently, lower values of UCS are obtained for WSA-C5 mixture.

5 CONCLUSIONS, RECOMMENDATIONS AND FUTURE INVESTIGATION

A series of UCS tests were carried out on WSA cement-based mixtures (WSA-C-L) and WSA-calcined kaolin mixtures (WSA-CK-L) that contained different cement and lime (or calcined kaolin) contents. Both cement plus lime or calcined kaolin contents as water demand W/B ratio varied to explore the optimum WSA recycling and compressive strength efficiency for different WSA mixtures. Some major conclusions were drawn as follows:

- (1) UCS values of WSA reference sample i.e. 100% WSA seems to slightly decreased along the 28 days curing time nearing 2 MPa.
- (2) WSA cement-based mixtures show another hardening trend during the 28 days of curing. UCS slightly increases with time. A mixture with a WSA-cement mass ratio of 95/5 (minimal cement content) developed a UCS of 2.5 MPa which was the highest value at 28 days.

- (3) Lime addition does not affect UCS strength. This can be explained by the presence of 51% of CaO in the WSA and the presence of free lime (3.8%).
- (4) Considering WSA calcined kaolin-based mixtures with CK low additions ($\leq 10\%$), UCS increases with curing time. We observed particularly with only 5% of calcined kaolin, a 9 MPa UCS value is achieved at 28 days.
- (5) A 5% CK addition has shown a clear different behaviour comparing to higher CK contents during hardening period.

To more eco-friendlily produce materials by recycling maximum of WSA and minimising the consumption of cement or calcined kaolin, various mixes with different volumes of WSA were investigated and conclusions above mentioned. But recycling 100% WSA without any addition does not seem acceptable for any material in terms of strength. So, two types of addition were proposed: a cement and calcined kaolin, to increase the strength of WSA mixtures.

The highlighted results of this study confirm that using of 5% of cement should be more suitable in non-structural construction materials or stabilized materials as a road sublayer with adding other acceptability tests. Concerning calcined kaolin as addition, a CK dosage of 5% could provide materials for applications in building materials. It is still necessary to investigate and to optimize this dosage.

The present study has revealed that the recycling of wastepaper sludge ash (WSA) as the main secondary raw material is clearly promising and possible. A more detailed research work will help to ensure better use of this waste material and for the most suitable recycling routes.

Based on the results obtained in present study and suggested recommendations, future research work will concern the following:

- (1) Durability behavior of wastepaper sludge ash-based materials to investigate the influence of severe climate conditions (wetting/drying, freeze/thaw) on mechanical strengths.
- (2) Environmental behavior during the curing time by analyzing the possible presence and amounts of heavy metals. Leaching tests are recommended on WSA raw materials and on the WSA-based mixtures. Both behavior studies could be coupled.
- (3) Full-scale behaviour of wastepaper sludge ash materials. A full-scale road sublayer will be implemented and some elements of building materials (paving units, bricks and cinder blocks...) be manufactured. So, WSA-based materials will be investigated in situ conditions. Scale effects between laboratory-site could be analyzed and discussed with technical difficulties observed during site applications.

6 ACKNOWLEDGMENTS

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