

Reuse of Paper Mill Ash in Plaster Blends

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Abstract: This work is part of a broad research conducted to study the re-use of industrial wastes in an economical and environmentally sustainable manner. Re-use of lightweight ash from the paper mill industry in the manufacture of plaster blends is examined in this study. Lightweight ash is produced during the combustion process of de-inking sludge. Studies demonstrate that lightweight ash is an excellent ingredient in the production of cement mortars and/or pozzolanic mortars to be used as plaster. In this research, ash valorization was researched by investigating the efficacy of the ash as it is, as a substitute for very fine sand.

If properly mixed at an optimal mix proportion, the mechanical strength ranges from 4-10MPa with specific gravity between 1150 and 1350 Kg/m³. Water absorption may be reduced to a very low level with silane-based surface treatment.

Keywords: Paper mill ash, de-inking ash, plaster, mortar, aggregate replacing, permeability, water absorption, compressive strength.

INTRODUCTION

This study was developed as part of a broader research aimed to demonstrate the technical, environmental and economical benefits of recycling an industrial by-product such as paper mill fly ash.

According to the European Commission's "Strategy on the prevention and recycling of waste" [1], the main thrust of their strategy lies in amending the legislation to improve its implementation and in preventing waste and promoting effective recycling. Parallel to these political aspects, setting out guidelines and describing measures aimed at reducing pressure on the environment caused by waste production and management, new researches and aspects of innovation must be pursued in order to put the strategy into practice.

Following this topic, the present paper deals with an industrial research that identifies new market applications for paper mill by-products as additives or ingredients in construction materials. Re-use of lightweight ash in the manufacture of plaster blends is examined in this study.

Lightweight ash comes from the combustion of paper mill sludge. In turn, the sludge is a by-product of the pulp and paper industry as well as paper recycling mills. If properly managed, paper by-products (such as ash and sludge) represent a typical example of industrial symbiosis [2] where all resources (materials, energy, water, etc...) fulfil a specific role in the industrial process. Once used, these materials should be salvaged and if necessary, modified or reprocessed to be reused elsewhere. As such, resources can be used again in other sectors (i.e., the construction sector). Despite the potential and the metaphor of the natural

eco-system [2], the eco-efficient utilization of pulp mill by-products presupposes to identify new final destinations that are more market attractive and environmentally friendly. Literature examples of industrial symbiosis applied to the overall processes of the paper mill industry are available in open journals and books.

In the Italian contest, where the paper industry has a long tradition and competitive clustering, de-inking sludge is an increasing as well as promising waste typology resulting from the separate collection of industrial solid waste [3, 4]. An average of 6 kg of sludge is produced from every ton of paper. In 2004, the total Italian production was about 6·10⁵ tons [5]. Historically, paper mill sludge has been used as landfill or as potential soil amendment and forest fertilizer [6-9]. The practice of employing fertilizer requires a better knowledge of and strict control over an undesired release of contaminants (especially metals) into the environment [6]. Only in few sporadic cases they have been used in the production of bricks [10-12].

On the other hand, landfill disposal has raised costs and difficulties in acquiring new sites due to environmental conservation regulations. Moreover, landfilling is a waste of resources because paper sludge is a readily recyclable product and useful material.

At the Center for By-Products Utilization of the University of Wisconsin-Milwaukee (Milwaukee, USA), researchers have proposed to reuse it in the manufacture of [13-15]:

- dry-cast concrete products such as masonry blocks and paving stones;
- wet-cast precast products;
- structural-grade concrete;
- and in controlled low-strength materials (CLSMs), also called flowable slurry or flowable fill, that are

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low-strength concrete-like material for non-structural use.

Other final destinations of paper mill sludge are in the paper industry itself when co-burnt with wood residue for producing heat and power for internal use. In that case, it could be more widely used as combustion material in a combined heat and power plant (CHP), according to the European Directives 2004/8 on Co-generation and 2008/98 on wastes [16]. These combustion residues are commonly classified as fly ashes as they come from the air emission control systems of the plants. The recent policy that sustains the use of bio-fuels (the 20-20-20 Europe targets [17]) is increasing the annual amount of fly ash produced in Europe, more than just paper industry.

According to existing experimentation, in the overall product life cycle, ashes from the combustion processes of paper mills are collected and recycled as a new by-product. They can be mostly reused as materials for the construction sector [18]. At the Institute National de Sciences Appliquées (INSA, Lyon, France), researchers have developed a calcination process to make them more pozzolanic and therefore more performing in concrete applications [19]. The calcination process increases the market value of ashes. Other attempts have been made to produce cements [20] and self-compacting concrete [21].

However, industrial applications of paper ash need further investigation and innovation in order to become more market attractive and environmentally friendly. According to the (revised) European Waste Framework Directive [22], the paper objectives follow the waste hierarchy and focus on both waste reduction and recycling. In fact, the study aims to:

- reduce the use of natural resources (like natural sand) and reduce the amount of waste in the paper mill industry by identifying new final destinations for wastes;
- recycle paper mill ash without any additional processing, that is without any additional cost.

Moreover, the Directive also aims to demonstrate that paper mill ash could be a performing ingredient in the production of mortars and plasters, although it is rather far from being an industrialized solution in its original state ("as it is").

The performance of interior/exterior plasters mainly depends on the properties of the mixed material (both sand and binder), the thickness of the layers and its application. Depending on the kind of application, it is possible to design a proper plaster by getting special properties based on the type of aggregates, admixtures and/or mineral additions used.

Frequently, the plaster selection is based on the external surface appearance; thus innovation in plaster design should be aimed to improve surface finishing. Paper ashes quite often exhibit lighter and finer properties as well as cheaper peculiarities than traditional ingredients: these properties should deal with improving surface finishing.

In this work, the ashes were provided by "Burgo Group" paper mills (Mantova, Italy). They were obtained from the combustion process of de-inking sludge. The initial

composition of the sludge was up to 55% of dry matter along with water. As far as the paper ash composition is concerned, it was mainly calcium silicates and oxides in addition to magnesium and aluminium oxides. They are classified as non-hazardous special waste and can therefore be disposed of in type B landfills according to Italian regulations [23, 24]. Similarly, such classification is expressed under EU and USA rules [21, 25].

MATERIALS AND METHODS

In the experimental stage, the following materials were used:

- paper mill ash;
- Portland cement type CEMII AL 42.5R (according to EN-197/1);
- limestone powder (calcium carbonate);
- natural sand with a maximum diameter of 2mm (according to EN 1097-6 and EN 933-1);
- and a silane-based hydrophobic product.

The paper ash, provided by "Burgo Group" paper mills (Mantova, Italy), comes from a traditional process of coated paper, more precisely from the combustion process of de-inking sludge. Paper wastes from the local productive plant were mostly employed in combustion. The main elements of the paper ash composition were:

- silica oxide (30-40%);
- calcium oxide (30-40%);
- aluminium and magnesium oxides.

The first phase involved the characterization of the paper ash in terms of its mineral composition (as mentioned above), bulk density of the dry powder, particle size distribution and surface analysis.

The bulk density of the paper ash, equal to 1200 kg/m³, was lighter than ordinary sand (that conventionally ranges from 2500 to 2600 kg/m³) and binders (2100-3100 kg/m³) generally used in the manufacture of mortars. Particle size distribution by using lazer granulometer (DigiSizer 5200) is reported in Fig. (1).

Ash particles were below 250µm, such as the finer fractions of sand; at least 50% of them were below 80 µm, which is the typical size of binders and fillers.

This fineness and lightness raise the use of paper ash as the only aggregate in plasters, promising a good surface finishing to plasters.

The shape and surface texture of particles were observed by a Scanning Electron Microscope (SEM).

Shape and texture were evaluated because of their influence on mortar mixture consistency and on water requirement. Results (see Fig. 2) detected high open particle porosity, which is the first indicator of the amount of water required to saturate it. The total water amount needed to be properly taken into account during the optimization of the mix design.

Mixtures of Portland cement - type CEMII AL 42.5R - lime and paper ash were prepared to reproduce plasters.

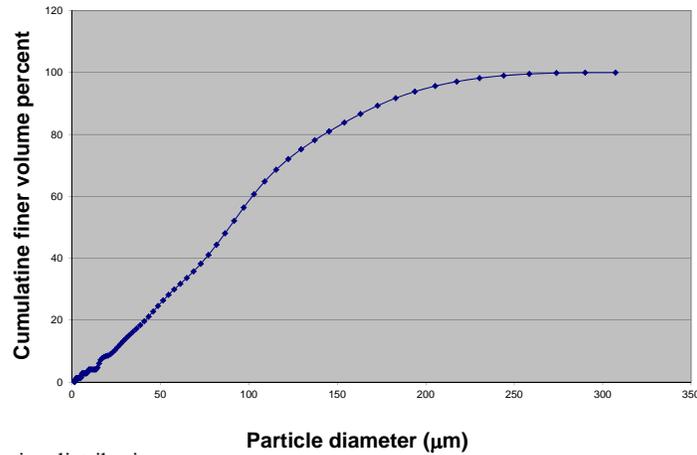


Fig. (1). Paper fly ash particle size distribution.

Table 1 shows the characteristics of the mixed mortars produced.

Several proportions of mixtures were made varying the ratio of paper ash and binder (pa/b in Table 1), namely M1, M2 and M3. In some mixture proportions, namely M4 and M5 (Table 1), natural sand was used to partly replace paper ash in order to obtain a proper mixture for several plaster types.

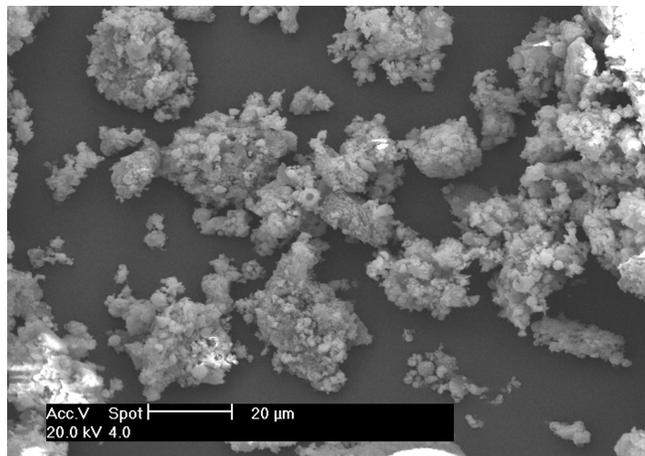
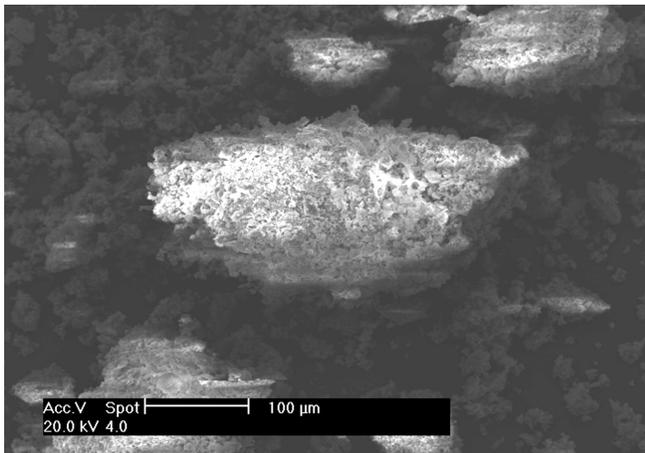


Fig. (2). Paper fly ash observed under a SEM at two different spots (20 μm and 100μm).

A conventional plaster mixture composition without paper ash or other inert materials (M0 in Table 1) were considered as reference in terms of water-binder ratio and specific weight.

Table 1. Characteristics of Mortar Mixes

Mortars	pa/b [-]	w/b [-]	sf [mm]	sw [Kg/m ³]
M0	-	0.61	145	1240
M1	1/1	1.7	150	1265
M2	2/1	2.27	135	1215
M3	3/1	3.5	125	1165
M4	1/1	1.56	140	1370
M5	1.5/1	2.1	145	1352
M6	2/1	2.24	140	1262
M7	2/0.5	2.20	130	1225
M8	3/0.5	3.16	135	1177

NOTE:

- pa/b is the paper ash/binder ratio by weight [-]
- w/b is the water/binder ratio by weight [-]
- sf is the slump flow in fresh mortars [mm]
- sw is the specific weight in mortars at 28 days of curing time [Kg/m³]

In all these mixtures (from M1 to M5), a conventional binder, made of lime and cement with equal weight (50% cement, 50% lime), was used.

In addition, carbon fly ash and calcium carbonate were used as partial substitution of the main binder. In these cases the composition was modified:

- the first consisted in replacing the cement with carbon fly ash (M6) - traditionally well known as material with good pozzolanic properties;
- the second consisted in replacing lime with limestone powder, i.e., calcium carbonate (M7, M8).

A hydrophobic product was used as surface treatment in some selected specimens (Table 3).

The water/binder (w/b) ratio reported in Table 1 is very high because of the fineness of the paper ash rate and because it comprises water “surplus”. Water surplus is defined as the water necessary to fully saturate the open porosity of paper ash as previously specified. This aspect is not negligible due to the high porosity of paper ash showed by SEM detection. The surplus estimated an average value of $40\% \pm 2$ by weight of dry paper ash.

The mortars were characterized in both fresh and hardened conditions by means of respectively the workability at the flow table (UNI EN 1015-3: 2000) and the mechanical properties at 28 days of curing time. Mechanical properties consist of compressive strength (UNI EN 1015-11: 2001).

Capillary water absorption (UNI EN 1015-18: 2004) and water vapor transmission (UNI EN 1015-19: 2008) were also measured in order to better characterize the most common properties of the plasters.

Experimental stage was performed on prismatic specimens (40x40x160mm). At least 2 samples for each mixture were tested, according to the standard requirement aforementioned. The values of the results represent the average of these multiple tests.

RESULTS AND DISCUSSION

The measured workability was quite scattered ranging between 125mm and 150mm (see figures in Table 1). To meet workability values, as recommended by the UNI-EN 1015 guidelines (slump flow ≥ 150 mm), small adjustments should be requested. As such, any adjustment (water addition or plasticizer admixture) was not made because the workability obtained was considered to be close to that required for most traditional applications (handcraft application). Moreover, the modification in the water/binder ratio led to a change in the performance of the hardened material. Alternatively, an aware amount of plasticizer admixture can be added to adjust only workability, but paying great attention to avoid bleeding effects. However, investigation in greater detail is necessary to find the best compromise.

The specific weight, less than 1300 Kg/m^3 , allows us to classify most of the mixtures created as light mortars.

The mechanical characteristics (see Table 2) turned out to be within a range of values that fit with almost all classes of resistance provided by traditional mortars. In fact, compressive strength values > 3.5 MPa are mostly used in several operative conditions. Recommended operative conditions versus compressive strength are available in the table of the UNI EN 998-1:2004 annex.

Scattering in the strength range could be mainly ascribed to several factors or to a combination of the following:

- the paper ash/binder ratio (which was changed as reported in Table 1, see M1, M2, M3);
- the partial replacement of paper ash with natural sand by 50% in weight (see in particular mortar M4 and M5 in Table 1);
- changing the binder (in M6 by replacing cement with carbon fly ash, mineral admixture with good

pozzolanic properties, while replacing lime with limestone powder, inert material, in M7 and M8).

In order to satisfy different mechanical requirements, it is certainly possible to extend the range of values obtained by making changes in the proportions of the mortars. This also fits with workability adjustment. According to the authors' suggestions, by adjusting the type of binder and the ratio paper ash/binder, the mechanical properties can joint to requirements. Similar conclusions have also been suggested by Fava *et al.* [26].

Water absorption coefficient (wa) was not negligible due to the high porosity of the mortars. It could be reduced to more negligible values by treating the surface of the mortar with hydrophobic products. In fact, a commercial silane hydrophobic product was used to treat the surface of some specimens after which the water absorption coefficient was newly calculated and later reported in Table 3. As expected, new water absorption values compared with those of Table 2, show a reduced water absorption coefficient. Therefore, such plasters with a negligible water absorption characteristic could be strongly recommended for external applications, i.e., surface exposed to weathering conditions. Because hydrophobic additive deteriorates with time, it should be periodically (indicatively every 4-5 years) renewed in order to avoid water absorption and plaster deterioration.

Table 2. Compressive Strength (Rc) and Water Absorption Coefficient (wa) of Mortars

Mortars	Rc [MPa]	wa [Kg/(m ² ·min ^{0.5})]
M1	9.5	3.6
M2	7	4.9
M3	5	7.5
M4	8	7.4
M5	7	5.8
M6	4	4.9
M7	5.5	9.5
M8	4	11.2

Table 3. Water Absorption Coefficient of Mortars without (wa) and with Hydrophobic (wa_h) Admixture

Mortars	wa [Kg/(m ² ·min ^{0.5})]	wa_h [Kg/(m ² ·min ^{0.5})]
M2	4.9	0.03
M3	7.5	0.09
M6	4.9	0.02
M7	9.5	0.08
M8	11.2	0.06

Results of water vapor transmission are showed in Figs. (3-5), namely mortars M2, M3, M6, M7 and M8. These mortars are characterized by the absence of natural sand and high paper ash content. The absence of natural sand simplifies the testing model (i.e., the transition area between

binder and inert is avoided) and enhances the effect of paper ash on water vapor transmission.

Fig. (3) compares water vapor transmission trends of two mortars having same binder (cement + lime) but different paper ash/binder ratio. Fig. (3) shows that the presence of a higher amount of paper ash increases vapor transmission, which is expressed as specimen weight loss by evaporation.

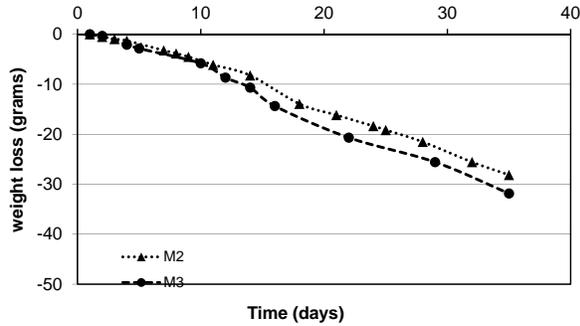


Fig. (3). Water vapor transmission in mortars having different paper ash/binder ratio (pa/b), in presence of cement + lime binder.

As previously outlined by capillary water absorption (see also water absorption coefficient in Table 3), it is confirmed that paper ash in mortars increases open porosity. Both high permeability and porosity drive in the characteristic of plaster breathability. On the other hand, also trends in Fig. (4) confirm the effect of the breathability of paper ash mortars. In this case, the binder is cement alone, and lime is replaced by limestone powder. The characteristic of breathability is mainly due to paper ash (see trend with higher paper ash and cement ratio) as any other ingredient is introduced into the specimen.

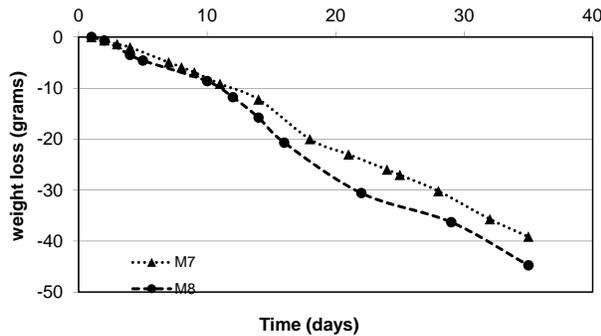


Fig. (4). Water vapor transmission in mortars having different paper ash/binder ratio (pa/b), in presence of cement alone.

Lastly, in Fig. (5), a comparison of the permeability of the mortars, prepared with the same amount of binder but of different nature, confirms the increased permeability of pozzolanic mortars (lime-carbon fly ash) compared to those with cement and lime.

As far as the water vapor transmission properties are concerned, their results are listed in Table 4. Values of water vapor transmission - expressed as resistance to vapor transmission (μ) - range between the low limits of 5 and 7. According to the table, the obtained values are quite close or point out only slightly differences. This is mainly due to the subsequent approximations in the numerical calculation of the water vapor transmission. In fact, a more revealing difference is showed in the Figs. (3-5). Despite that, the μ

values are clearly below the "threshold" set by European regulation for mortars. The European standard (UNI EN 1015-19: 2000) requires μ value ≤ 15 [-] when used as plasters on damp walls. However, it is advisable for mortars to show similar values, or slightly lower, than the masonry under the plaster. Nevertheless, plasters having high permeability to water are suitable in situations where it is essential to ensure breathability to the walls.

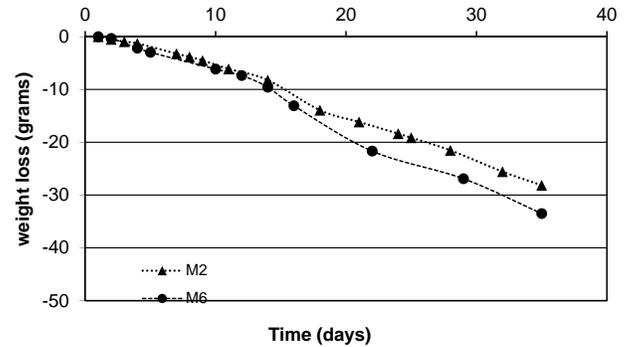


Fig. (5). Water vapor transmission in mortars having the same amount of paper ash/binder ratio (pa/b) but different binders (cement and lime: M2, lime and carbon fly ash: M6).

Table 4. Resistance to Water Vapor Transmission of the Mortars (μ)

Mortars	μ [-]
M2	7
M3	7
M6	6
M7	5
M8	5

CONCLUSIONS

The present paper focuses on the feasible use of paper ash from paper mills in the construction material sector. In particular, paper ash has been tested as an ingredient in the production of blends for internal or external plaster. For example, in finishing plaster, paper ash can be used as partial or whole replacement of the inert.

The best feature of this material is its lightweight property that gives fresh mortar proper consistency and facilitates its application both by hand (trowel) and by plaster machines. The final product will have low to medium density according to the percentage of the paper ash used in the mixture.

Resistance to vapor transmission, usually a relevant requirement for plasters, is clearly below the "threshold" level set by the European regulation for mortars. According to the values obtained, although slightly scattered, it appears that permeability is supported by the increasing of the paper ash.

The high absorption of water by capillary action, which frequently constitutes a problem in outdoor use, can be modified by treatment with water repellent products.

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CONFLICT OF INTEREST

Declared none.

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