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Use of MSWI Fly Ash for the Production of Lightweight Artificial Aggregate by Means of Innovative Cold Bonding Pelletization Technique. Chemical and Morphological Characterization

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Cement-based cold bonding pelletization process has recently gained relevant consideration in literature mostly due its suitability for the manufacture of artificial aggregates. An extensive study on the recovery of municipal solid waste incinerator fly ash is investigated. Such material is classified as hazardous and it needs a treatment to be used or landfilled. The examined fly ash from waste incineration plant were subjected to cold bonding pelletization process by using cement, lime and coal fly ash as binders to achieve their stabilization/solidification. Several mixtures were prepared employing different weight amounts of ash from 50% to 70%. A further pelletization step was performed using only cementitious binders in order to reach satisfactory immobilization levels. Lightweight porous aggregates with good properties in terms of density, water absorption and crushing strength were obtained from this process. The results consider these materials suitable for the recovery in the field of building materials. The same process was also tested on concrete specimens manufactured with the same aggregates. The results showed a lightweight concrete having average performance and mostly suitable for applications where high performance are not necessary.

1. Introduction

Industrial solid wastes represent a crucial concern and their contaminant potential, due to different pollution factors, is a widespread threat around the world. The specific treatment of industrial solid wastes plays a key role in order to favor a safer disposal and maximize the efficiency of possible recycling processes (Singh et al.2017).

Cold bonding pelletization of waste is often proposed as stabilization/solidification technique for waste reuse in low cost building materials production (Colangelo et al. 2017). Particularly, recycled artificial aggregates are of course one of the most interesting technological solution for waste recovery (Baykal et al. 2000. Cioffi et al. 2011. Colangelo et al. 2013). Cement-based cold bonding pelletization process has recently received a quite relevant attention in literature and its suitability for the manufacture of artificial aggregates is undoubtedly worthy of consideration (Alunno et al. 2010. Margallo et al. 2014. Gesoglu et al. 2012. Gomathi et al. 2015. Di Palma et al. 2012. Vasugi et al. 2014. Di Palma et al. 2015). In the present study a stabilization/solidification process based on cold bonding pelletization which makes use of a rotary plate pelletization pilot-scale apparatus with binding mixes is investigated.

In addition to the traditional single step pelletization, a double-step pelletization is proposed to have final products with improved properties (Colangelo 2015). This process has been applied to several mixes in which the waste content has been varied from a minimum of 50% (wt. %) up to a maximum 70%.

In the one-step pelletization the waste is incorporated within the binding matrix in a measure ranging from 50 to 70%. In the two-step pelletization a second step is carried out with pure binder to get the aggregates from

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the one-step process encapsulated within an outer shell able to improve the technological and leaching properties.
Such approach has been extensively tested in the laboratory through the production of recycled aggregates using a rotary plate pelletization pilot-scale apparatus. Such approach has economic and environmental advantages due to the reduced energy requirement (process carried out at room temperature) respect to the industrial alternatives such as sintering (Ferone 2013. Galiano 2011), which is an energy intensive process. Thermal processes produce high quantity of CO_2 and it is very difficult to obtain environmental permit for industrial scale plant. More recently, alternative cement-free binding matrices with reduced embedded CO_2 have been proposed for stabilization/solidification (Zheng 2011. Zhou et al.2017) such as geopolymer and alkali activated ones. These systems have gained an increasing interest from researchers thanks to promising results in terms of mechanical, physical, durability properties and possibility of synthesis starting from natural/industrial wastes (Colangelo 2016. Kourti 2010) for a wide range of applications (Lampris 2009. Messina 2015. Molino 2014. Messina 2013. Ferone 2015). A further reason of interest in cold bonding pelletization is that this technique, if applied on an industrial scale, above all in developing countries, the so-called BRICS, could allow a significant reduction of quarrying activities, which determine a relevant depletion of natural resources, with an associated irreversible impact on landscapes (Tang et al. 2017). The replacement of natural aggregates with artificial ones could help to reduce the related environmental impact and address waste management towards materials recovery and resource efficiency (Coppola et al. 2016. Dehdezi et al.2015. La Rosa et al. 2016. Kumar et al. 2017).
In this study a stabilization/solidification treatment based on cementitious cold bonding pelletization has been employed in order to manufacture artificial aggregates by using a rotary plate pelletization pilot-scale. We used municipal solid waste incinerator (MSWI) fly ash samples and cement, lime and coal fly as binders. In order to avoid that chlorides and sulfates contained in MSWI fly ash could affect the cementitious binding matrix, a pre-washing treatment has been carried out to reduce their content (Colangelo 2012. Shi 2017).
In the present process a second step pelletization after the traditional single step has been performed to obtained final products with enhanced properties. In the one-step pelletization 50%-70% of the waste is mixed with the binding matrix while in the two-step pelletization a pure binder is used to encapsulate the aggregates coming from the one step within an outer shell. This further step has proved to be very effective to improve the technological and leaching properties. An artificial aggregate obtained by double step pellettization is illustrated in Figure 1.

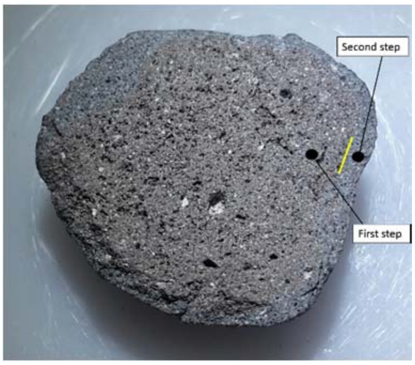

Figure 1: Artificial aggregate obtained by double-step pelletization.

2. Materials and methods

The fly ash employed in the present study comes from an Italian incineration plant located in Melfi (Potenza, Italy) that treats municipal, hospital and industrial wastes. it is equipped with rotary and stoker furnaces. This kind of waste is classified as hazardous materials according to the European Waste Catalogue, it contains heavy metals that have been determined through X-ray fluorescence and inductively coupled plasma atomic emission spectrometry (ICP-AES) technique. The granules are been manufactured employing as binder CEM II/A-L 42.5R (UNI EN 197-1: 2011), a commercial hydrated lime and coal fly ash supplied by the ENEL (Italian Electricity Board) power plant located in Brindisi (Italy). Since the cementitious matrix has a reduced capability of immobilize chlorides and other soluble salts, a pre-washing treatment has been done. In particular, the two examined MSWI fly ash samples have been submitted to a two-step washing pre-treatment with liquid/solid ratio equal to 2:1. as extensively described in (Colangelo 2012). Such liquid/solid ratio is an optimized process oriented to reduce soluble salts content and the production of liquid waste. In this study, the examined MSWI fly ash samples are divided in two halves and one of them has been treated with a 4:1 liquid to solid ratio. Then the mix is filtered and the recovered liquid is treated with the other halve for the second step of the washing pre-treatment. Each step lasts 30 minutes and the level of extraction reached is about 90%.

2.1 Aggregates and concrete manufacturing and characterization

The MSWI fly ash subjected to washing pre-treatment have been employed to prepare four pelletization mixtures, the compositions are reported in Table 1. The granules have been obtained using a pilot scale granulator apparatus having a rotating and tilting plate with a diameter of 80 cm. During the process, the rotating speed has been set at 45 rpm and at 45° for the tilting angle, according to optimized parameters reported in Colangelo et al. 2012. The granules obtained have been cured in a climatic chamber for 12 hours with a temperature of 50°C and a relative humidity of 95%. Such phase is very effective for the granules since it gives them the necessary hardening to be used for the handling phase. Then, the granules have been cured for 14 days at room temperature and humidity.
The four mixtures listed in Table 1 have been subjected to a second step using cement/coal fly ash as binder in the proportion of 40% by weight of granules.
Concrete mixtures have been obtained by varying the content of acrylic additives based on water absorption of the artificial aggregates in order to obtain the same machinability of the blends. The mixtures have been designed employing the produced cold bonded aggregates as coarse fraction (4-18 mm) and natural sand as fine fraction (0-4 mm).
Such mixes have been tested in order to evaluate compressive strength and dynamic modulus of elasticity.
Concrete cubic specimens having a dimension of 15 cm have been realized to measure the compressive strength, according to UNI EN 12390-3:2009 standard while cylindrical specimens having a diameter of 15 cm and a height of 30cm have been manufactured to determine the dynamic modulus of elasticity through an ultrasonic pulse measurement.

Table 1: Composition of the matrices (wt%).

Mix	Binder composition			
	CEM	Lime	CFA	w/S
R70C	30	-	-	0.25
R70L	-	30	-	0.25
R60LA	-	15	25	0.32
R50LA	-	30	20	0.35

3. Experimental results

Detailed images on the interface and on the different internal and external porosity have been obtained using SEM microscope (Figure 2). In the first photo (a), the protective shell is highlighted and it can be noted that it should be improved in the next studies to avoid the formation of macrovoids. The second photo (b) shows a magnification of the matrix and it is noticeably compact. All the spheres shown in the figure b are the ash from incinerator. In the pictures on the right (c,d) a macrovoid is shown. Such macrovoids are formed inside the granules. It can be also noted that within the macrovoids it is possible to observe the hydration products of concrete. In fact the space presents into the granules, allows cement hydration products to expand. Such phenomenon results in agitated shapes as shown in figure. Future work will deal the micromechanical study on artificial aggregates and matrix in composite materials (Fantilli et al. 2017. Khezrzadeh 2017. Talò et al. 2017).

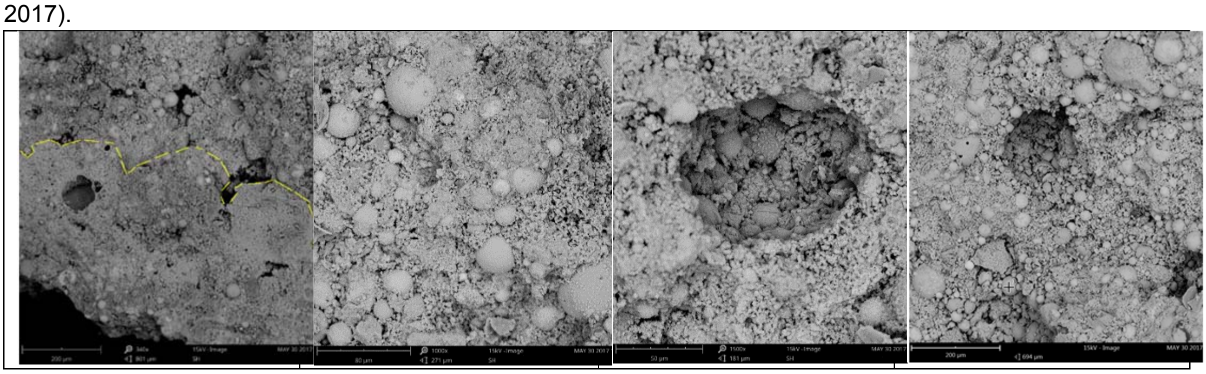

Figure 2: SEM images of artificial aggregate showing the matrix and the macrovoids.

EDS analysis has been performed on the produced artificial aggregates. Such test is very good at determining the elements present in samples. The results have been plotted with X-ray wavelength on the X-axis and intensity on the Y-axis and each peak has been labeled with its corresponding element (Figure 3).

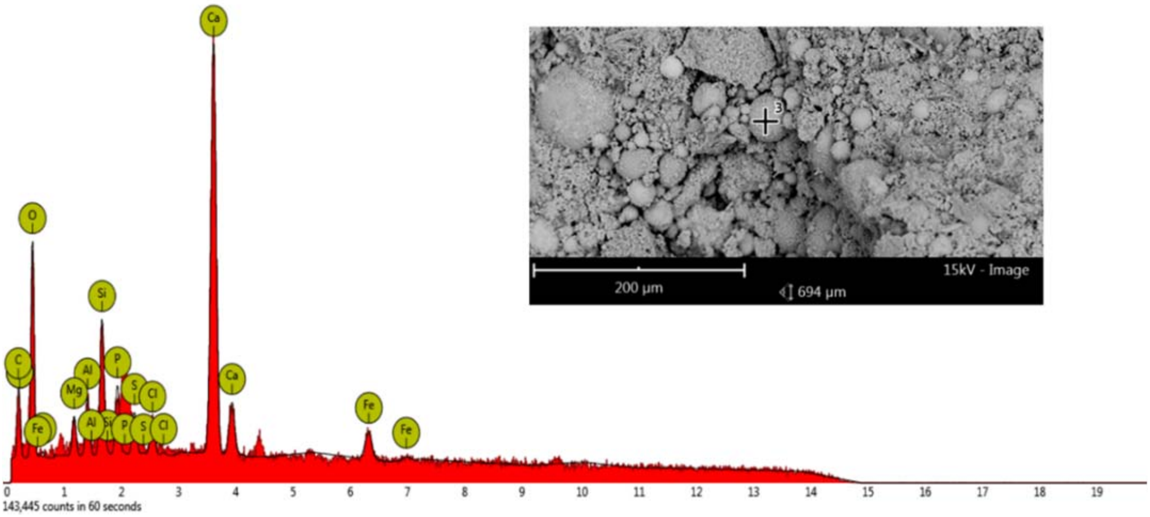

Figure 3: EDS test performed on the produced artificial aggregates.

Table 2: Chemical composition by EDS test

Element Number	Element Symbol	Element Name	Atomic Concentration
20	Ca	Calcium	12.4
8	O	Oxygen	46.8
14	Si	Silicon	3.0
15	P	Phosphorus	1.7
13	Al	Aluminium	1.7
6	C	Carbon	29.0
16	S	Sulfur	1.0
26	Fe	Iron	2.3
17	Cl	Chlorine	0.8
12	Mg	Magnesium	1.4

4. Conclusions

The experimental data discussed in the previous paragraphs, lead to the following conclusions:

· in the production of cold bonded artificial aggregate the MSWI fly ash deriving from municipal, hospital and industrial solid wastes incineration can be recycled up to 70% content.

· the double-step pelletization process is able to enhance the physical, the mechanical properties and also the stabilization properties of the binding matrices. The employed approach forms a granule encapsulated in an outer layer made of waste-free binder that is capable to reduce the amount of heavy metal, the porosity and moreover it enhances the physical and mechanical properties.

· the most effective binders resulted cement and coal fly ash/lime systems even if different binders can be employed to manufacture the granules.

· comparing the two kinds of MSWI fly ash employed in this approach, the results highlighted that the one coming from the stoker produces granules with enhanced technological properties.

· the release of heavy metals in the most of the cases was found lower than the limits fixed by the Italian law for applications in civil engineering. A different behaviour has been noted by employing lime as a binder for the manufacture of the granules.

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