TREATMENT AND REUSE OF ASH FROM MUNICIPAL SOLID WASTE INCINERATION

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ABSTRACT

Incineration is considered one of the most convenient treatment of urban solid waste (MSW) as it allows a significant volume reduction and an energy enhancement of the waste itself. However, it cannot be considered a final treatment solution because of the formation of solid residues, mainly composed of two groups of ash, bottom ash (BA) and fly ash (FA). Their characteristics depend on the type of incoming waste and on the combustion methods. BA are considered non-hazardous waste, while FA, due to their high content of heavy metals, alkali chlorides and soluble metal salts, have the characteristics of hazardous waste. Among the various recovery possibilities is the use of FA for the production of artificial lightweight aggregates (LWA), used for the production of lightweight concrete (LWC). This article aims to highlight how the use of FA granules as aggregates in LWC can give good results in terms of compressive strength, rupture and elastic modulus. In fact, the particle size distribution and chemical composition of the FA, as well as the generally spherical shape and low cost, make this type of ash an ideal material for this use.

Keywords: incineration ash, lightweight aggregates, granulation, lightweight concretes.

1 INTRODUCTION

The use of artificial aggregates obtained from waste materials and by-products, as an alternative to natural aggregates, has aroused considerable research interest. Many industrial wastes, including soil waste, fly ash from municipal solid waste incineration (MSWI-FA), blast furnace slag (GGBFS) and marble sludge (MS), can be applied to make lightweight aggregates [1].

Municipal solid waste incineration ash (MSWI-FA), for example, which contains heavy metals, chlorides and sulphates potentially harmful to cement materials and human health, requires pre-treatment before it can be safely disposed of in landfills or recycled in the construction sector [2].

Extensive research was also conducted to pre-treat different types of waste to be used as raw materials for the production of artificial aggregates. Artificial aggregates can be produced by two types of processes: cold granulation and high temperature sintering. The cold granulation process has recently received quite significant attention in the literature which is rich in applications where many waste materials have been shown to have the potential to be used as feedstock: fly ash from combustors and municipal solid waste incinerators, metallurgical slag, furnace dust, sediment, shredding waste. Their suitability for the production of artificial aggregates is undoubtedly worthy of consideration. It is known that waste treatment is largely based on cement-based stabilization/solidification processes, which allow for safer disposal and/or recovery of materials for the manufacture of building materials [3].

These systems have shown promising results in terms of physical, mechanical and resistance properties and the possibility of synthesis starting from industrial waste, favoring economic benefits both in the field of waste cycle management, such as the reduction of material to be disposed of in landfills, and concrete ecological and energy advantages.

Therefore, in this paper, a study on the recycling of fly ash (FA) by the cold granulation process is proposed.

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2 MATERIALS AND METHODS

The fly ash (FA) used is filter residue produced by the treatment of fumes from the municipal solid waste incineration plant in Acerra (Naples). They are classified as hazardous waste in the European Waste Catalog, and cannot be used or even landfilled without prior treatment [4]. Therefore, a washing pre-treatment was carried out on FA in order to mainly reduce the content of heavy metals, chlorides and sulphates at the end of using the FA in the granulation process, which can be seriously compromised by the high content of soluble salts. For this purpose, the pre-treatment was carried out by washing in two phases with deionized water, each of 1.5 and led to a reduction of soluble salts, specifically a reduction of chlorides equal to about 67% and of sulphates 25%. After pre-treatment, the pre-treated fly ash was collected through a filtration process, and dried at 105 ± 5°C in an oven for 24 h.

In the present work, the granulation technique was performed to stabilize FA, using various mix-designs of a ternary blend for the production of lightweight artificial aggregates. The production of the granules was carried out by means of a granulator on a pilot scale (Fig. 1), equipped with a rotating and tilting plate (d = 80 cm) for which the rotation speed and the angle of inclination were respectively set at 45 rpm for the rotation speed and 45 for the angle of inclination.

Figure 1: Granulator.

During the single granulation process, different mixtures are made, in which the FA is present at 80%, while the mass of limestone cement and granulated blast furnace slag vary respectively by 15%, 10% and 5%. In addition to the traditional single-pass granulation, a second granulation step was carried out. In the single-pass granulation the waste is incorporated in a binder matrix, in the double granulation on the other hand, the second phase is performed with 70% of cementitious binder, to obtain encapsulated aggregates inside an outer shell, to improve the technological properties and of leaching, in order to reach satisfactory levels of immobilization [5], [6]. Three different mixtures (A, B, C) were made for the production of single granulation granules (Granules (I)):

1. Granules A (I): FA 80%, CEM 5%, GGBFS 15%;
2. Granules B (I): FA 80%, CEM 10%, GGBFS 10%;
3. Granules C (I): FA 80%, CEM 15%, GGBFS 5%.

The double granulation process was performed starting from the aggregates obtained from the single granulation process with the aid of a new mixture composed of 30% limestone cement (CEM II/AL 42.5R) and 70% marble sludge (MS) in order to increase the amount of reused waste (marble sludge) and increase the thickness of the aggregates, previously made. In the double granulation process, the granules obtained from the single granulation are subjected to double granulation (Granules (II)):

1. Granules A (II): Granules A (I); CEM 30%; MS 70%;
2. Granules B (II): Granules B (I); CEM 30%; MS 70%;
3. Granules C (II): Granules C (I); CEM 30%; MS 70%.

The light artificial aggregates obtained from the granulation process (Fig. 2) were cured for 28 days at room temperature. This curing phase favors the hardening of the granules, which is necessary for subsequent manipulations and in such a way that it significantly improves their technological properties. All the systems listed underwent a characterization in terms of physico-chemical and mechanical properties.

Figure 2: Single granulation granules (Granules (I)).

3 RESULTS AND DISCUSSIONS

3.1 Chemical-physical-mineralogical characterization of the precursors

The granulation process uses fly ash (FA), granulated blast furnace slag (GGBFS) and limestone cement (CEM II/A-L 42.5R) as components of the systems, for the production of lightweight artificial aggregates. In addition, an innovative additional granulation phase was performed with cement binder and marble sludge (MS) in order to achieve satisfactory immobilization levels. The fly ash, granulated blast furnace slag, limestone cement and marble sludge before being used in the granulation process, were characterized from a chemical-physical-mineralogical point of view, using the following analytical techniques: X-ray fluorescence (XRF), X-ray diffraction (XRD), scanning electron microscopy (SEM) and particle size analysis.

The chemical composition of the FA as such and of each constituent of the mixtures, that is, treated FA, GGBFS, MS and cement was determined by X-ray fluorescence, and reported in Table 1.

Table 1: Chemical composition in terms of equivalent oxides (wt%) of fly ash as they are (FA TQ), treated fly ash (FA), granulated blast furnace slag (GGBFS), marble sludge (MS) and limestone cement (CEM II/A-L 42.5R).

Chemical composition (wt%)	FA TQ	FA	GGBFS	MS	CEM
Fe_2O_3	0.86	1.16	25.53	1.35	3.41
CaO	24.31	39.90	17.48	51.92	67.16
CO	16.35	14.13	11.29	22.74	–
SiO_2	2.62	4.57	16.26	14.16	16.65
Al_2O_3	1.53	3.36	8.93	4.56	4.21
SO_3	8.57	9.58	–	–	5.34
MgO	1.09	2.57	7.94	1.21	1.71
Mn_2O_3	–	–	3.44	–	–
Cr_2O_3	–	–	1.84	–	–
NO_x	–	–	10.07	–	–
SnO_2	–	–	–	2.20	–
Na_2O	13.87	9.10	–	0.86	–
K_2O	6.41	2.32	–	1.02	1.54
TiO_2	0.36	0.98	–	–	–
ClO	21.20	8.77	–	–	–
ZnO	2.85	3.36	–	–	–

The examination of the chemical analysis was a fundamental step for understanding the chemical matrix of the precursor, in fact, from the result of the X-ray fluorescence analysis (XRF), of the fly ash as such, it is possible to notice a presence of CaO higher than the other elements, in particular with respect to the SiO_2 and Al_2O_3 content. This abundance may be, in the first instance, due to the type of waste that was fed to the combustion chamber and, moreover, linked to the presence of Ca in the plant, since calcium-based materials are used in acid gas abatement systems. Also significant are the fraction of chlorine and sulphur equal to approximately 21% and 9% respectively. From the XRF analysis of the treated fly ash, it appears that the pre-treatments play a fundamental role by potentially reducing the soluble salts, and in particular the presence of chlorine which is approximately 9%.

Furthermore, it was possible, by means of X-ray diffraction analysis (XRD), to identify in the FA as they are, the presence of crystalline species containing calcium, such as, Lime (CaO), Calcite ($CaCO_3$), Portlandite ($Ca\ (OH)_2$), Calcium sulphate ($Ca\ (SO)_4$), and chlorine-containing phases such as sodium chloride and potassium chloride (NaCl and KCl), calcium chloride hydrate CaClOH. The identification of these crystalline species strengthens the XRF chemical analysis, in which a high percentage of calcium and chlorine is found compared to the other elements. Another crystalline species easily distinguishable from the diffraction spectrum is SiO_2. The treated fly ash has some crystalline phases found in fly ash as such as, Calcite ($CaCO_3$), Portlandite ($Ca\ (OH)_2$) and Calcium sulphate ($Ca\ (SO)_4$), in addition there are also other hydrated crystalline species containing calcium such as Syngenite ($K_2Ca\ (SO_4)_2{\cdot}H_2O$), Gypsum ($CaSO_4{\cdot}2H_2O$), Calcium chloride dehydrate (Ca $(ClO)_2 \cdot 3H_2O$). The presence of these last mineralogical phases is justifiable given the pre-treatment of the ashes, carried out by washing with water. Another difference that can be seen from the mineralogical analysis of fly ash as it is and those treated is that in the latter there are fewer crystalline phases containing chlorine (NaCl).

SEM micrographs were made for the samples of fly ash and treated fly ash, which showed the presence of a rough and irregular surface. Finally, a particle size analysis was carried out. Table 2 shows the particle size distribution of the fly ash, from which it can be seen that the particles of the treated FA are larger than the untreated ones.

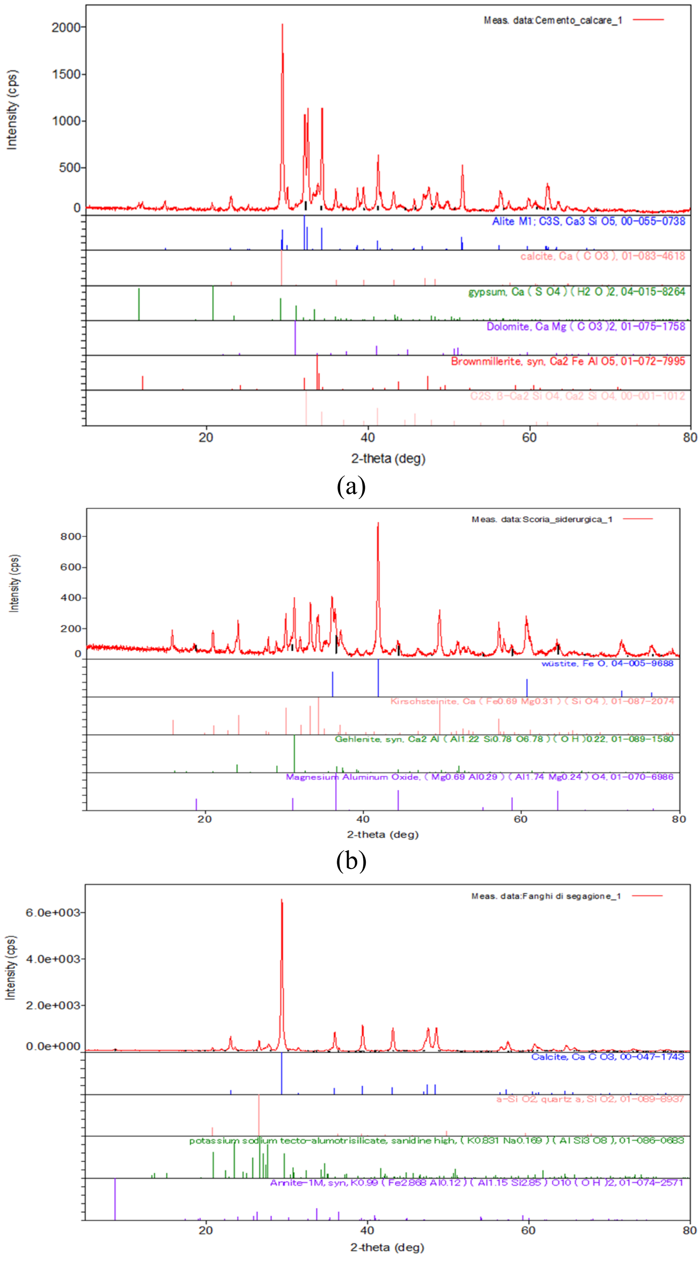
Table 2: Particle size distribution of fly ash as they are (FA TQ), treated fly ash (FA).

Granules distribution (%)	FA TQ	FA
<1 µm	0	0
1–10.5 µm	5.84	4.75
10–20 µm	9.28	6.30
20–49 µm	29.35	16.73
49–80 µm	13.66	8.89
80–120 µm	8.29	6.03
>120 µm	33.58	57.30

The remaining components of the mixture, that is, GGBFS, MS and cement, in addition to being characterized by X-ray fluorescence (XRF), whose compositions in terms of equivalent oxides are reported in Table 1, were also characterized by diffraction analysis X-ray (XRD), scanning electron microscopy (SEM) and particle size analysis.

The results of the XRD analysis of limestone cement show that the main crystalline phase is Alite (Ca_3SiO_5) which gives the material a development of its resistance in the short term; followed by Belite (Ca_2SiO_4), which performs a function similar to halite but acting over the long term. Furthermore, these calcium silicates, reacting with water, form Portlandite, a crystalline phase with a Ca $(OH)_2$ structure, and an amorphous phase of hydrated calcium silicate, commonly known as C–S–H, responsible for setting and hardening of the paste. There are further crystalline phases such as Calcite ($CaCO_3$), Gypsum ($CaSO_4$ $2H_2O$), Dolomite (CaMg $(CO_3)_2$) and Brownmillerite (phase containing iron and aluminium Ca_2 (FeAl) $2O_5$). To ensure the principle of identity for the mineralogical components, even the blast furnace slag (GGBFS), waste produced by the steel industry, was analyzed using the X-ray diffraction (XRD) method. The mineralogical phases present in GGBFS are Wustite (FeO), Monticellite (Ca (FeMg) SiO_4), Gehlenite (Ca_2Al (AlSiO) OH). Finally, the crystalline phases present in the marble sludge (MS) are Quartz (SiO_2), Calcite ($CaCO_3$), ((KNa) (ASi_3O_8)) and Annite ($KFeAlSi_3O_{10}$ $(OH)_2$). Fig. 3 shows respectively the diffractogram and the mineralogical components detected in the cement, blast furnace slag (GGBFS) and marble sludge (MS).

The electronic scanning micrographs were also made on concrete, blast furnace slag and marble sludge. SEM micrographs of the cement at higher magnifications show the presence of particles with a rough and irregular surface and the presence of crystalline phases. The slag from the blast furnace has a rough and porous surface where it is possible to see a very extensive amorphous matrix and a minor presence of crystalline phases. For the marble sludge it is possible to note how the sample has a fine particle distribution, micrographs at higher magnifications show the presence here too of a rough and irregular surface, and the presence of crystalline phases, as shown in Fig. 4.

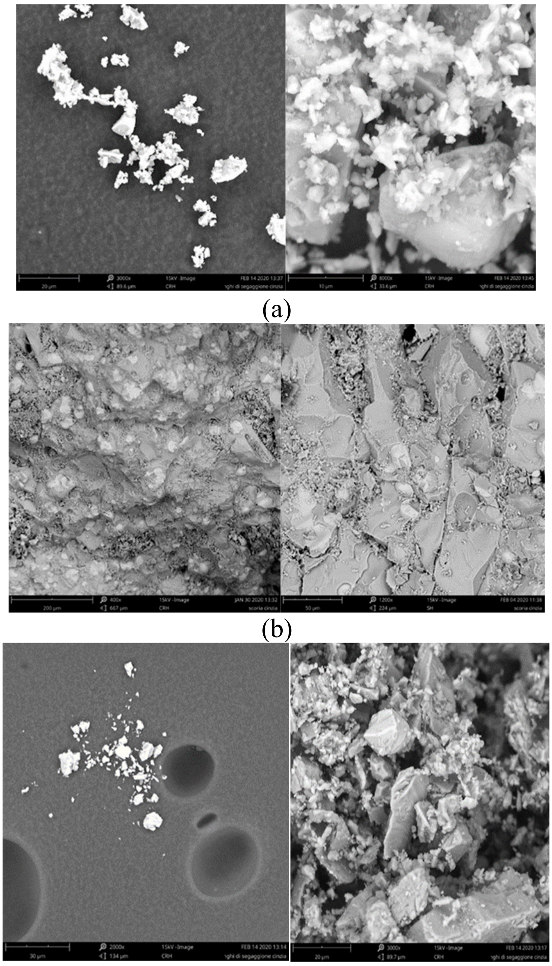


(a)

(b)

(c)

Figure 3: X-ray diffraction analysis (the reflection angle 2θ on the x axis and the intensity on the y axis) of (a) limestone cement (CEM II/A-L 42.5R); (b) granulated blast furnace slag (GGBFS); and (c) marble sludge (MS).



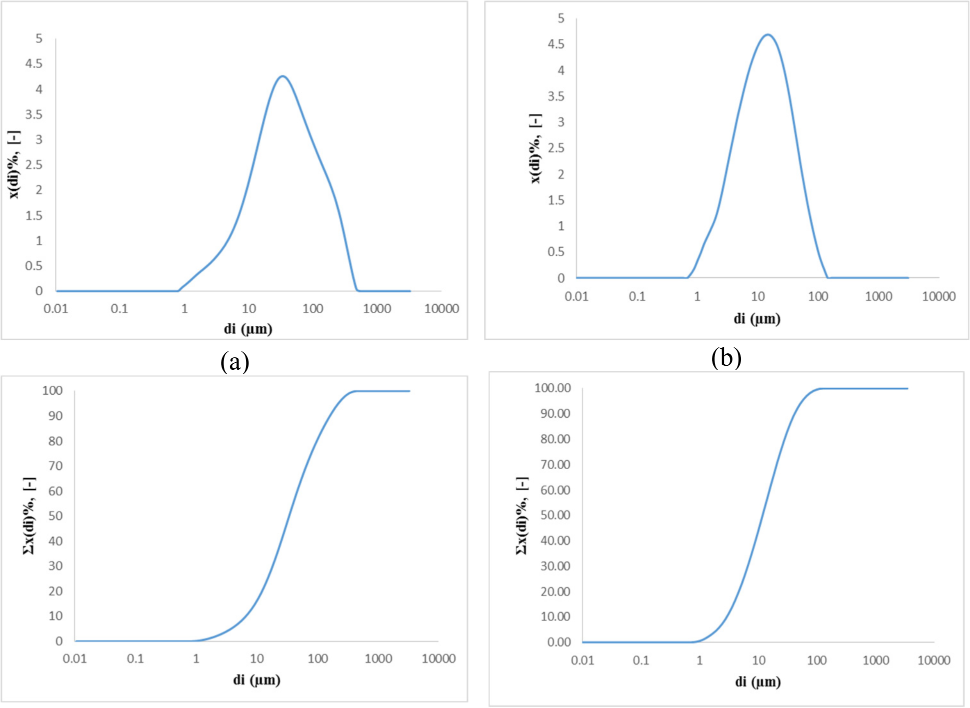
(a)

(b)

(c)

Figure 4: Scanning electron microscopy of (a) limestone cement (CEM II/A-L 42.5R); (b) granulated blast furnace slag (GGBFS); and (c) marble sludge (MS).

Among the numerous factors influencing the granulation process, the initial particle size distribution is one of the most important properties of the raw material. In this regard, the absolute particle size distribution of blast furnace slag (GGBFS) and marble sludge (MS) is reported. GGBFS exhibits a monomodal absolute particle size distribution and the representative diameter of the particle population is 33.2 µm. The marble sludge appears to have a monomodal particle size distribution and the representative diameter of the particle population is 15.5 µm. In addition to the absolute particle size distribution, Fig. 5 also shows the cumulative particle size distribution of GGBFS and MS respectively. From the cumulative particle size distribution, it is possible to deduce another characteristic parameter, namely the median, respectively for GGBFS d50 = 33.15 µm and for MS d50 = 13.5 µm.



(a) (b)

(c) (d)

Figure 5: Absolute particle size distribution (on x axis the values of the weight fraction of each particle size $x(di)$ and on y axis the values of the particle diameter di) of (a) granulated blast furnace slag (GGBFS); (b) marble sludge (MS) and cumulative particle size distribution (on x axis the sum of the values of the weight fraction of each particle size $x(di)$ and on y axis the values of the particle diameter di) of (c) granulated blast furnace slag (GGBFS); and (d) marble sludge (MS).

3.2 Chemical-physical and mechanical characterization of light artificial aggregates

3.2.1 Particle size analysis

After 28 days of curing, the light artificial aggregates obtained by a single and double granulation process were characterized in terms of physic-chemical and mechanical properties. Granules with dimensions ranging from 2 to 20 mm were obtained. The particle size distribution of the granules was determined according to the procedure described in the UNI EN 933-1 standard. In order to determine the particle size distribution of Granules A (I), Granules B (I), Granules C (I), Granules A (II), Granules B (II), Granules C (II) respectively, a sieving was carried out (or screening), a method used to establish the particle size distribution of a granular solid. The particle size analysis by sieving was performed by means of special sieves arranged in series, in the case in question of 2 mm, 4 mm, 8 mm, 10 mm, 16 mm, 20 mm, each of which retains the fraction of solid whose granules they are larger than the sieve holes. As regards the particle size distribution of Granules A (I), there is a higher concentration of granules passing between the sieves with an opening of 4 mm, 10 mm and 16 mm. Results similar to the previous case were also observed for Granules B (I). Finally, also for Granules C (I) it is possible to note that there is a higher concentration of granules passing between the sieves with an opening of 8 mm and 16 mm, including the sieve with an opening of 10 mm. Specifically, a greater presence of granules with dimensions greater than 10 mm of opening of the sieve was observed. Furthermore, only for Granules C (I), there are no granules having a diameter equal to 2 mm. This factor could be caused by the greater quantity of binder used, resulting in better agglomeration of the powders compared to the other two mixtures and the formation of granules with larger diameters. As regards the particle size distribution of granules A (II), in general, the percentage of the different diameters is well distributed. In particular, granules having dimensions greater than 16 mm of opening of the sieve, appear to be present in greater quantities, compared to the other granulometric classes. Furthermore, comparing these results with those obtained for Granules A (I), a substantial increase in the average size of the diameters is observed, in accordance with the single granulation process. Finally, from the results of the particle size distribution for Granules B (II) and Granules C (II), it is possible to make the same considerations made for Granules B (I) and Granules C (I), i.e. that there is a quantity greater than granules with a diameter of 16 mm and that substantially the rest of the particle size is well distributed.

3.2.2 Impact test

The impact test was performed on lightweight artificial aggregates, which is used to determine the impact factor of the aggregates and select them for a specific application. The impact test, in accordance with the UNI 12620-4 standard, consists in introducing the granules for a particle size class between 10–14 mm in a cylindrical container (d = 10 cm), which fills up to half. The apparatus consists of a metal sliding rod ended with a round point of 1.6 mm diameter, mounted in a suitable frame. The cylinder containing the granules is positioned inside the press, the piston is dropped 15 times, and a load of 8.9 kN is applied to the test sample. The test sample is collected and extracted from the cylinder. Using a 2 mm sieve, proceed to pass all the material below this dimension and proceed with the measurement of the weight determining the percentage of passage. Below are the results of the impact test carried out on the granules made in this report (Table 3); it should be remembered that this test allows to determine the percentage in heap of the granules which serves to define the final destination of the granular aggregates. In particular, according to the quantity of material passed through the 2 mm sieve, they are classified as: <15%, extremely strong; between 15% and 45%, satisfactory for road paving; >45%, extremely weak.

Table 3: Impact test results on single granulation granules (Granules (I)) and double granulation granules (Granules (II)).

	Percentage passing through the 2 mm sieve (%)
Granules A (I)	30.56
Granules B (I)	22.22
Granules C (I)	25.00
Granules A (II)	19.44
Granules B (II)	25.00
Granules C (II)	44.44

As can be seen from the results shown in Table 3, all the granules are satisfactory for use as a filling for road pavement, according to the UNI 12620-4 standard. In principle, the impact resistance is higher in Granules C (II), i.e. granules with 15% cement, 5% blast furnace slag and 80% FA, with the further addition of 30% cement and 70% sludge.

3.2.3 Compression test

With the compression test, the compressive strength which represents the maximum value of the applied stress is determined. In this study, compression tests were carried out on granular aggregates, in accordance with the UNI EN 12390-3 standard, using a Controls hydraulic press from Matest, on single and double granules with a diameter of 12.5 mm. Below are the results of the compression test on the granules, carried out in this report (Table 4).

Table 4: Press test results on single granulation granules (Granules (I)) and double granulation granules (Granules (II)).

	(MPa)
Granules A (I)	1.33
Granules B (I)	1.45
Granules C (I)	1.86
Granules A (II)	1.95
Granules B (II)	5.36
Granules C (II)	10.94

From the results of the granules, obtained from the single granulation process, it emerges that the breaking load, on the other hand, is almost constant, varying from a minimum of 1.33 MPa to a maximum of 1.86 MPa for the granules obtained from the single granulation process. The results of the granules obtained from the double granulation process are even more satisfactory than the aggregates obtained from the single granulation. In fact, by comparing all the values of the breaking loads, it is evident that the double granulation aggregates all have higher breaking loads than the corresponding aggregates obtained from the single granulation. This is because the granules obtained from the double granulation process have a greater quantity of binder, or 30% more by weight of cement, therefore, the second granulation phase determines an increase in compressive strength. As a result, the double granulation is more effective, in terms of mechanical performance, than the single granulation.

3.2.4 Assignment test

Release tests are tests in which a solid material is placed in contact with a liquid (leaching agent), resulting in a liquid product (eluate). The chemical analysis of the eluate allows to determine the chemical species that are released from the solid material over time, also contextualizing the potential danger to the environment. These tests were carried out for the granules, following the procedures described in the UNI 10802 standard. The samples were placed in contact with demineralized water, in liquid/solid ratio = 10, and left under stirring for 24 h. The solid residue was then separated by filtration, and the eluate obtained was analyzed on the atomic absorption spectrometer (AAS) for the determination of heavy metals. Furthermore, the leaching solution was analyzed by ionic liquid chromatography for the determination of chlorides and sulphates. Table 5 shows the results relating to the release of heavy metals, chlorides and sulphates (in mg L-1) of the granules deriving from the single and double granulation process and the relative limit values for non-hazardous waste.

Table 5: Results of the release test for single granulation granules (Granules (I)) and double granulation granules (Granules (II)).

	Cu	Pb	Zn	Cd	Cr	Chloride	Sulphates
Granules A (I)	0	1.15	1.11	0.01	0.45	39,439	7,283
Granules B (I)	0	0	0.28	0.04	0.1	22,930	5,158
Granules C (I)	0	0.71	0.32	0.06	0.55	22,996	14,859
Granules A (II)	0	0.49	0.62	0.03	0.49	9,819	4,601
Granules B (II)	0	0	0.17	0.01	0.35	7,101	909
Granules C (II)	0	0.24	0.29	0	0.53	13,333	3,595
Law limits[a]	5	1	5	0.1	1	1,500	2,000

[a] UNI 10802.

As for the results relating to the concentration of heavy metals, the only value that does not comply with the legislation is the concentration of Lead for Granules A (I). However, it should be noted that the value deviates slightly from the regulatory limit. Conversely, the leaching values of chlorides and sulphates were beyond the limits for almost all granules, except for Granules B (II) where the resulting sulphate concentration was below the limit. Also in this case, the double granulation process has shown benefits and it can certainly be said that the new conferment of thickness is able to better trap contaminants.

4 CONCLUSIONS

The irreversible and increasingly worrying crisis, together with environmental destruction, has led researchers to seek alternative solutions to natural resources. In particular, the aim is to promote the use of recycled aggregates to protect and support natural resources. Many studies have mainly confirmed the feasibility and efficiency of using recycled aggregates as building materials to reconsider the appropriateness of reusing industrial waste. In this regard, this study was conducted, from which it is possible to state that the light artificial aggregates produced by double granulation have brought numerous environmental and economic advantages, with the addition of a further waste (marble sludge) to give the new thickness. This translates into lower cost of waste disposal, lower cost of product preparation and less space in landfills. The results in terms of mechanical properties, deriving from this study, demonstrate the effectiveness of using various wastes in the production of aggregates through the double granulation process. At the same time, the results of the leaching tests showed a significant global release of the soluble salts. However, leaching from granules obtained by double granulation was reduced for both chlorides and sulphates.

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