

# WET GRANULATION FOR INERTIZATION OF FLY ASHES, "FLUFF", AND MARINE SEDIMENTS

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## Abstract

Some experimental results on the inertization of fly ashes from coal thermal plants (CFA), the non-metallic fraction of automotive shredder residues (ASR or "fluff") and dredged marine sediments (MS) are reported. A series of mixtures, containing hydrated lime or cement as binder, with the aim to obtain pastes with a satisfactory compression strength and to optimize the percentage of granulated wastes, are formulated utilizing the wet granulation technique at room temperature.

Inertization with hydrated lime is fit for treatment of fly ashes, while cement is more suitable for stabilization of the other two residues, in order to obtain light weight aggregates for concrete production.

**Key words:** Granulation, coal fly ashes, automotive shredder residues (ASR), marine sediments, metals, leaching, aggregate.

## 1. Introduction

The main effort in the future, according to the European directives on wastes [1], will be devoted to the disposal of only inertized and incinerated wastes in controlled landfills. So that in a near future a huge quantity of wastes must be treated utilizing different technologies.

First effort will be devoted to the individualization of wastes, then, the development of new and cheaper technologies for the recycling and reusing will be very interesting both from an economical and ecological point of view. In this work three different types of dangerous and impactable wastes are selected and experimented (coal fly ashes [2,3], "fluff" [4,5] and marine sediments [6,7]).

Fly ashes from coal thermoelectric plant could have a great environmental impact because of the large amount of production (39 Mt/y in European Community), so that they must be reused. In case of coal fly ashes the reuse and the recycling could be practically complete, owing their pozzolanic behaviour, in fact until now, cement and concrete industry can easily absorb the whole production of these ashes.

Every year in UE nations about 12 M vehicles are shredded, utilizing particular treatment methods, 75% of components of the vehicles can be recycled, while the remaining part (25%) constitutes a waste [4]. So that around 3 Mt/y of automotives shredder residues (ASR) are generated: half of ASR is composed by rubber, glass or plastics, which can be either transformed into alternative fuel or recycled, the second half, typical "fluff", is incombustible and it has not yet been valorised.

Harbour basin and waterways have to be dredged in order to maintain an adequate shipping channel depth. The obtained sediment is de-watered in drainage pounds and the deposited in a landfill for contaminated harbour sediment [6,7].

Authors think that a cheap and efficient method to reuse these wastes is inertization by means of wet granulation at room temperature. So that the aim of this paper is to characterize these residues and to investigate the physical and mechanical behaviour of inertized products with the final goal to produce a light weight aggregate that will be utilize to produce concrete [2,3].

## 2. Materials and methods

### 2.1 Materials

Experimental tests were performed on fly ashes (silico aluminous product) obtained by the thermoelectric plant "Enel" located in Brindisi (Italy), "fluff" in produced in an automotive shredding plant "Italferro" at S. Palomba (Roma, Italy), marine sediments derive from the harbour of Piombino (Livorno, Italy).

In the granulation tests calcium hydroxide  $\text{Ca}(\text{OH})_2$  and a Portland cement CEM II-A/LL 42.5 were used as binders; in order to decrease water demand during the mixing a commercial superplasticizer (BASF ACE 363) was utilized.

Table 1 shows the chemical composition and the heavy metals contents of the selected materials: all data were assumed by previous works [2,4,8].

Component	CFA (%w)	ASR (%w)	MS (%w)	Component	ASR (mg/kg)	MS (mg/kg)
$\text{SiO}_2$	46.5	28.8	63	Cadmium	11	< 1
$\text{Al}_2\text{O}_3$	24.4	5.3	11	Chromium	< 2	32
$\text{Fe}_2\text{O}_3$	10.1	25.6	8.3	Copper	3727	50
CaO	7.0	8.1	1.8	Lead	7420	27
MgO	1.1	2.9	0.9	Zinc	450	65
$\text{Na}_2\text{O}+\text{K}_2\text{O}$	1.8	2.7	2.5	Residue 105 °C (%w)	91.1	68.9
LOI (1100 °C)	5.2	14.0	9.6	Residue 600 °C (%w)	39.4	-

**Table 1:** Chemical composition (oxides) and heavy metal contents of selected materials

### 2.2 Experimental procedures and analyses

The aim of experimentation is to set up a process to produce aggregate for concrete utilized the previously mentioned wastes. Proposed process deals with three steps: the selection of the product, the granulation and the preparation of concrete samples.

The first step of experimentation had the aiming at selecting a fraction characterized by a negligible heat of combustion and a grading range to be suitable to be granulated in the subsequent phase.

Coal fly ashes were utilized as produced, while plastics and foam materials were separated by grinding the ASR produced in the plant and passing a 4 mm diameter mesh, marine sediments are crushed without any further treatment. Granulation was performed in a sloping tank rotating at 22 rpm equipped with four mixing paddles, granulation tests were performed using a total amount of mixing of 1 kg in each test.

A commercial hydrated lime was utilized as binder for fly ashes to get a general idea of the pozzolanic activity of the ashes, while cement (CEM II-A/LL 42.5) was utilized to treat “fluff” and marine sediments.

The influence of following parameters on the possibility and range of granulation and on granules diameter was evaluated: 1) water content; 2) waste/binder ratio and 3) addition of a superplasticizer.

The experimental conditions of the first series (series I) of tests are summarized in table 2.

Test	Binder (L)	Superplasticizer	CFA/L	W/(CFA + L)
I.1	Lime	no	9	0.4
I.2	Lime	no	4	0.4
I.3	Lime	no	2.3	0.4

**Table 2:** Experimental conditions in the granulation of fly ashes

The experimental conditions of the second series (series II) of tests are summarized in table 3: in all tests the ratio between binder ( C ) and the sum of coal fly ashes (CFA) and “fluff” (F) was maintained equal to 0.3, while water dosage was the operating parameters. Superplasticizer was added at 1.9 (%w) and 3.8 (%w) with respect of the sum of cement an fly ashes.

Test	Binder ( C )	Superplasticizer	F/CFA	F/C	C/(F+CFA)
II.1	Cement	no	1	1.7	0.3
II.2	Cement	yes (1.9 %w)	1	1.7	0.3
II.3	Cement	yes (3.8 %w)	1	1.7	0.3

**Table 3:** Experimental conditions in the granulation of “fluff”

The experimental conditions of the third series (series III) of tests are summarized in table 4: all experiments were carried out utilizing the same operative conditions of the previous series of tests (series II).

Test	Binder ( C )	Superplaticizer	MS/CFA	MS/C	C/(MS+CFA)
III.1	Cement	no	1	1.7	0.3

**Table 4:** Experimental conditions in the granulation of marine sediments

After a 28 days period of curing at ambient temperature in moisture saturated room, the granules were subjected to compressive strength tests, according to the UNI EN 13055 Part I [9], and leaching tests, according to Italian regulation [10]. Specific weight of the granules was measured by hydrostatic weighing.

A Philips PU 9200 atomic absorption spectrophotometer was used to measure the metals content in the leached solutions.

### 2.3 Composition of the concrete mixes

The granules produced in the second series were then used as a coarse aggregates in concrete samples, prepared according to the UNI 11013 [11]. The mix design of the samples is shown in table 5, all the tests were performed in triplicate. The reference aggregate A1 was a calcareous one.

Sample number	A1	A2
Cement (CEM II 42.5) (g/l)	350	350
Water (g/l)	175	175
Aggregate (g/l)	1325	550
Siliceous sand (Torre del Lago) (g/l)	500	500
Water retention after 30 min (g)	6	113

**Table 5:** Mix design of concrete samples

## 3. Experimental results and discussion

### 3.1 Granulation tests

Figures 1, 2 and 3 show the experimental results of the granulation tests for series II; in particular figure 1 shows the results of the tests carried out without the use of a superplasticizer (s.p.), while figure 2 and 3 show results of tests carried out, at the same experimental conditions, utilizing a superplasticizer.

Results show that increasing the water content, with respect the whole weight of the mix, the average granules diameter increases, in fact in steady growth system, the granules growth rate increase rapidly with increasing water content [12]. Moreover utilizing a superplasticizer at the same average granules diameter (2 mm) water dosage decreases.

The test performed on marine sediments shows that, at the operating conditions investigated, granulation allows the production of almost spherical shaped granules, but only about 60% of the total amount of powder charged in the granulator at the beginning of the tests was converted into granules. The remaining part constitutes a sludge at the bottom of the tank.

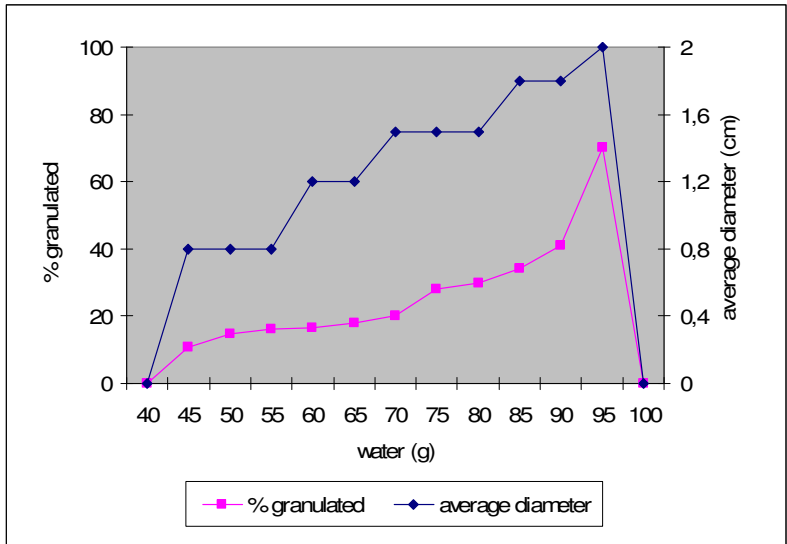


Figure 1: Granulation test (series II) without s.p.

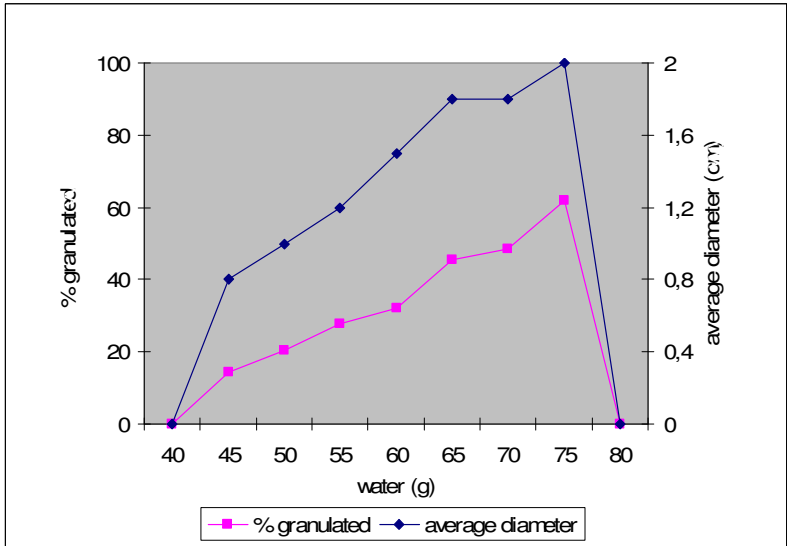


Figure 2: Granulation test (series II) with s.p. (1.9 %w)

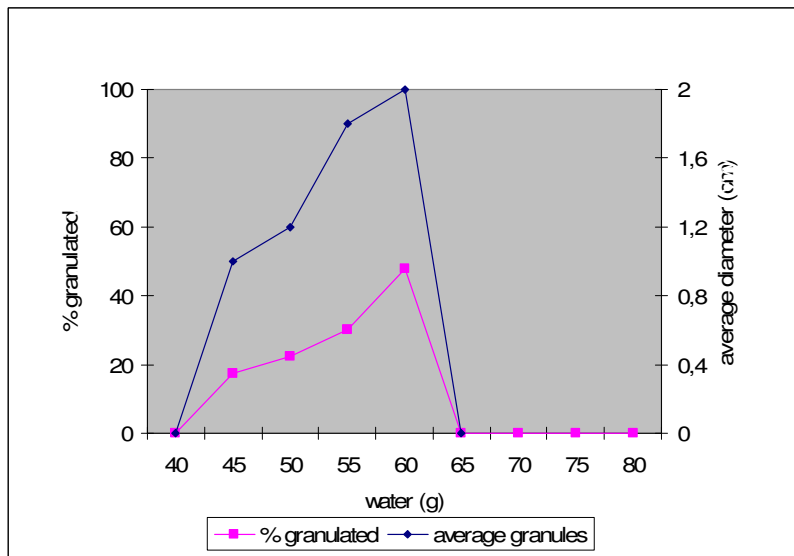


Figure 3: Granulation test (series II) with s.p. (3.8 %w)

### 3.2 Compressive strength tests

Table 6 shows the results of the compressive tests on the granules in the three series of tests, in the same table is reported the granulometric range of produced granules .

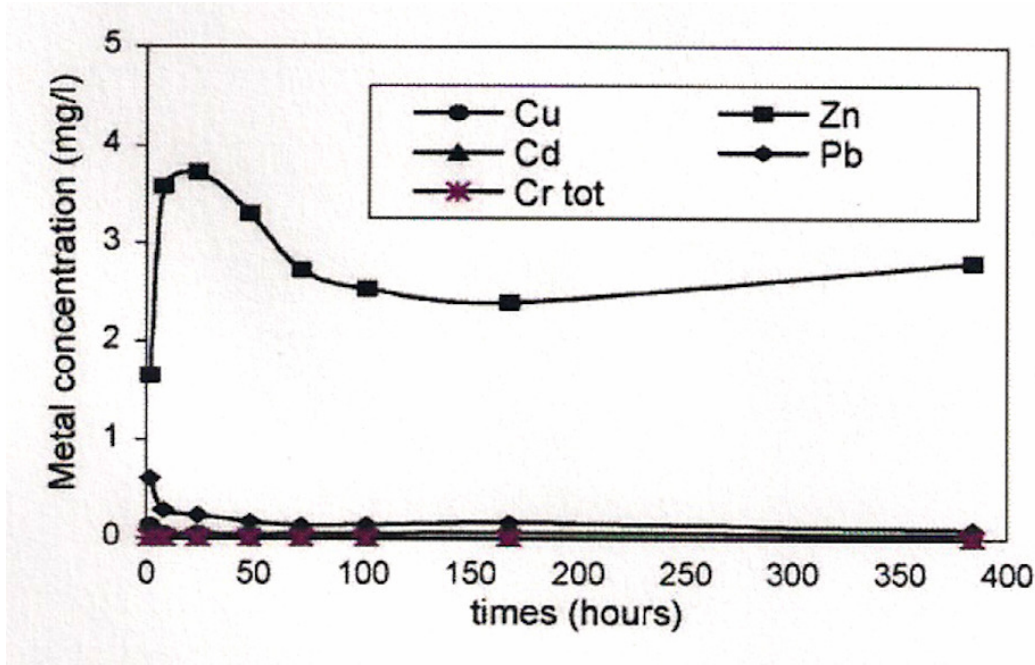
	Specific weight (kg/m <sup>3</sup> )	Compressive strength (Mpa)	Granules diameter (cm)
Series I	1150	4 - 9	0.8 - 1.2
Series II	1400	1.2 - 1.5	0.3 - 2.0
Series III	1350	0.4 - 0.8	0.8 - 2.0

**Table 6:** Compressive strength on the produced granules

The obtained values of compressive strength for the concrete samples prepared, using selected granules (series II) as aggregate, are 25.8 and 14.3 (MPa) for samples A1 and A2 respectively [13].

### 3.3 Leaching test

Figures 4 show the results of the leaching tests performed on the granules of the series II. Figure show that the only metal that is heavily released in the environment is zinc, exceeding the Italian threshold limit (3 mg/l). In fact it is well that zinc interact with the cement clinker grains during hydration and retard cement setting: this effect may explain the long term release of the zinc [14].



**Figure 4:** Leaching test performed on produced granules series II (ASR)

In the case of marine sediments granulation, the total absence of metals in the leachate from the produced granules, confirms the immobilization action towards hazardous substances, as already observed in other immobilization studies [14,15].

#### 4. Conclusions

In this paper the possibility to reuse industrial sub-products like coal fly ashes, the non-metallic residues of an automotive shredder residues and dredged marine sediments was investigated. Such wastes was stabilized utilizing the wet granulation technique to produce light aggregates for cementitious mixes.

Process of granulation/stabilization, naturally, requires a preventive selection and characterization of the waste materials.

Coal fly ash may be directly utilized to produce granules: experimental tests showed that, utilizing a mix of (5-10) %w of hydrated lime and (90-95) %w of fly ashes , may be produced granules with a compressive strength (4-9) MPa and a specific weight equal to 1150 kg/m<sup>3</sup>.

Instead "fluff" and marine sediments must be pre treated and sieved to reuse the fraction under 4 mm mesh. To stabilize these wastes cement, as binder, was employed: experimental tests pointed out the maximum waste (either "fluff" or marine sediment) (W)/fly ash (CFA) ratio and the optimal cement ( C)/waste (W) + fly ash (CFA) ratio. Ratios  $W/(CFA) = 1$  and  $C/(W+CFA) = 0.3$  allow to obtain granules in a range (3-20) mm of diameter and up to 1400 kg/m<sup>3</sup> of specific weight.

Moreover experimental data showed that the size of the produced granules is a function of water content in the mix, in other words water addition controls the process and the rate of granules growth. It is a consequence that utilization of superplasticizers allows to obtain the same average granules diameter diminishing water dosage in the mixes. Leaching tests showed, for all wastes, a good level of immobilization of heavy metals, except for zinc.

Finally concrete samples prepared using the produced granules as aggregate showed a specific weight up to 1800 kg/m<sup>3</sup> and a compressive strength up to about 14 MPa.

Synthetically this work allowed to determine maximum capacity to stabilize wastes (W/CFA) and the optimal dosage of binder (C/W+CFA), while further investigation are needed to know the influence of other parameters like water viscosity, water and binder content on granules growth.

Authors think that wet granulation of industrial wastes is nearly an "empirical art", so that a significant and growing quantitative understanding on wastes granulation processes is needed.

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