

# Production of aggregate from non-metallic automotive shredder residues

Vito Alunno Rossetti, Luca Di Palma\*, Franco Medici

*Dipartimento di Ingegneria Chimica, dei Materiali, delle Materie Prime, Metallurgia, Università di Roma "La Sapienza",  
Via Eudossiana 18, 00184 Roma, Italy*

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## Abstract

In this paper, the results of an experimentation on the production of granules suitable to be used as aggregates in cementitious or asphalt mixes are presented and discussed. The granules were obtained by granulating the non-metallic fraction of automotive shredder residues.

In a preliminary separation step the fluff fraction containing mainly inert and non-metallic materials was sieved and analyzed for the metal content.

In the following granulation step, the sieved fraction was mixed with binding materials, fly ash and a densifier agent, to produce granules of 5–30 mm of diameter and up to 1400 kg/m<sup>3</sup> of specific weight. The granulation was carried out at room temperature in a rotating tank.

Concrete samples prepared using as aggregates the produced granules showed a specific weight up to 1800 kg/m<sup>3</sup> and a compressive strength up to about 55% of reference samples prepared using a calcareous aggregate, depending on the fluff content of the mixes, and on the nature of the binder and of the other components used.

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## 1. Introduction

Every year, in the UE about 12 M vehicles are shredded, and 8 or 9 Mt/year of wastes are produced: this amount will probably increase in the next future, as a consequence of the continuous expansion of the automotive industry. To minimize waste production, there is need to develop new strategies of management of the whole life cycle of the vehicles [1].

Right now two possible recovery pathways are outlined: the reuse of parts or components of the vehicle, and the separation and reuse of the metal fraction (iron). This second pathway generally involves the shredding of the vehicles and a ferromagnetic separation of the ferrous fraction. This treatment produces a large amount of residues, the so-called fluff (ASR, automobile shredder residues) that constitutes about 25 wt.% of a vehicle. Fluff contains rubber, glass, plastics, polyurethane foams, wood, textiles, paints, adhesives and non-ferrous metals, and is generally disposed in landfill as hazardous waste. As a consequence, until today, about 25 wt.% of an end-life vehicle is landfilled,

and this may cause the contamination of soil and groundwater. Those residues, in fact, may contain significant amount of hazardous substances, such as heavy metals, polychlorobiphenyls (PCB), chlorofluorocarbons (CFC), together with a number of both organic and inorganic compounds.

Moreover, this type of wastes, produced at a rate of about 3 Mt/year, represents up to 10% of the whole amount of hazardous wastes produced per year in UE, and about 60% of the total shredding wastes [2].

In this context, two EU directives have recently changed the legislative body on vehicle disposal. The EU Directive 2000/53/CE (about end life vehicles) stated that the vehicle recovery and recycling in term of weight percentage must compliance the 85% of the whole vehicle before January 2006 and the 95% before January 2015 [3].

At the same time tighter requirement have been fixed for landfill disposal of urban and industrial waste, in particular for the biodegradable and combustible fractions, according to the EU Directive 1999/31/CE [4].

As a consequence of both the increasing amount of automotive shredder residues production, and the tightening of Environmental Regulation requirement for landfill disposal, a new effort is necessary to found an alternative pathway for recycling

\* Corresponding author. Tel.: +39 06 44585571; fax: +39 06 44585622.  
E-mail address: luca.dipalma@uniroma1.it (L. Di Palma).

and reusing the fluff fraction. In effect, at the moment, there is not any fully developed technology for this purpose.

A promising solution could involve the industry of concrete production considering its increasing difficulties in finding natural aggregates as a result of the increasing use along the years of rock and mineral. This has addressed the concrete producers to alternative aggregate sources, such as synthetic ores, and, even, some treated solid wastes.

Several studies have been performed to evaluate the possibility to reuse wastes in the production of aggregate for the concrete industry, eventually through an intermediate treatment step. Processes aiming at recycling construction wastes [5,6], metallic wastes [7,8], mining wastes [9], municipal incinerator wastes [10,11], dredged sediments [12,13] and other types of wastes [14,15], are already fully developed.

Previous experimentations [16,17] have assessed that residues from end-life vehicles can be used in the production of aggregates for concrete, provided a preliminary transformation into a suitable product. This can be realized by a thermal treatment, or at room temperature.

In this paper, a process to use part of the non-metallic fraction of automobile shredder residues as aggregate for concrete and asphalt mixes is proposed.

The process set-up involved the immobilization of raw fluff in granules produced at room temperature, by mixing selected amount of fluff with a binder in the presence of other additives. The granules were then used as aggregate in concrete samples that were compared with common light aggregates in term of physical and mechanical properties.

The immobilizing effectiveness towards heavy metals, ions and organic matter was also evaluated, performing leaching tests both on granules and concrete samples.

## 2. Materials and methods

### 2.1. Materials

Experimental tests were performed on the fluff produced in the Automotive Shredding Plant "Italferr" at S. Palomba, Roma, Italy. In that plant, up to about 150 t/day of vehicles are treated and about 35 t of fluff are daily produced.

In the granulation tests calcium hydroxide,  $\text{Ca}(\text{OH})_2$  or CEM II-A/LL 42.5 were used as binders. In order to increase the cohesion of the pellets to get the granulation, a densifier (an aqueous emulsion of an acrylic polymer, POLICRIL VHT, F.A.R., Milano, Italy) was added to the mixing water.

To enhance mechanical properties of the granules also the addition of fly ash produced in the thermoelectric plant of Brindisi, Italy, was investigated.

Table 1 shows the composition and selected characteristics of the fly ash used.

### 2.2. Experimental procedures and analysis

The aim of the experimentation was to set up a process to produce aggregate for concrete from the non-metallic fraction of the automotive shredder residues.

Table 1  
Fly ash composition and characteristics

| Total composition                                     |         | Water soluble fraction (4.7%) |         |
|---|---------|-------------------------------|---------|
| Component   | % (w/w) | Component                     | % (w/w) |
| $\text{SiO}_2$  | 46.5    | CaO                           | 32.2    |
| $\text{Al}_2\text{O}_3$                               | 24.4    | MgO                           | 2.6     |
| $\text{Fe}_2\text{O}_3$                               | 10.1    | $\text{CO}_2$                 | 0.7     |
| CaO   | 7.0     | $\text{SO}_3$                 | 52.7    |
| MgO   | 1.1     | $\text{Cl}^-$                 | 0.3     |
| $\text{Na}_2\text{O} + \text{K}_2\text{O}$            | 1.8     | $\text{OH}^-$                 | 2.5     |
| $\text{SO}_3$   | 1.5     |                               |         |
| L.O.I. (1100 °C)                                      | 5.2     |                               |         |
| Specific surface (Blaine) 6800 $\text{cm}^2/\text{g}$ |         |                               |         |

The proposed process deals with three steps: the selection of the product, the granulation, the preparation of concrete samples.

The first step of the experimentation had the aiming at selecting a fraction characterized by a negligible heat of combustion and a grading range suitable to be granulated in the subsequent phase. Plastics and foam materials were separated by grinding the fluff produced in the plant and passing a 4 mm diameter mesh, while iron residues were separated with a magnetic system.

Granulation was performed in a sloping tank rotating at 22 rpm equipped with four mixing paddles, as shown in Fig. 1. Granulation tests were performed using a total amount of mixture of 1 kg in each tests.

The influence of the following parameters on the possibility and range of granulation and on granules diameter was evaluated:

- water content;
- fluff/binder ratio;
- other components content.



Fig. 1. Granulation tank.

Table 2  
Experimental conditions in the granulation tests

| Series | Binder (C)       | Densifier (D) | F/FA ratio | F/C ratio | FA/C ratio |
|--------|------------------|---------------|------------|-----------|------------|
| I a    | Lime             | Yes           | 0.333      | 1.25      | 3.75       |
| I b    | Lime             | Yes           | 1          | 2.5       | 2.5        |
| II a   | Lime             | No            | 0          | 0         | 5          |
| II b   | Lime             | No            | 0.333      | 1.25      | 3.75       |
| II c   | Lime             | No            | 1          | 2.5       | 2.5        |
| II d   | Lime             | No            | 3          | 3.5       | 1.5        |
| III a  | CEM II-A/LL 42.5 | No            | 0          | 0         | 5          |
| III b  | CEM II-A/LL 42.5 | No            | 0.333      | 1.25      | 3.75       |
| III c  | CEM II-A/LL 42.5 | No            | 1          | 2.5       | 2.5        |
| III d  | CEM II-A/LL 42.5 | No            | 3          | 3.5       | 3.5        |

Three series of experimental tests were performed: in the first series lime was used as binder together with fly ash and the densifier as additives; second series the same tests were carried out without the densifier; third series of tests the binder was a CEM II-A/LL 42.5.

The experimental conditions of the tests are summarized in Table 2: in all the tests the ratio between binder (C) and the sum of fly ash (FA) and fluff (F) was 0.2: fluff (F) content was the operating parameter. Densifier was added at 2 wt.% with respect to lime content.

Each series consists of a family of tests that differ from one another in water content. Water dosage was varied to individuate a range from a minimum necessary to obtain the granules, to a maximum corresponding to the formation of a semifluid sludge in the granulator (granulation range).

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 1 [18], and leaching tests, according to Italian Regulation [19]. Specific weight of the granules was measured by hydrostatic weighing.

The pH of the leached solutions was measured with a Crison 421 pH meter; a ionic chromatograph Dionex DX-120 was used to determine ionic species; a Philips PU 9200 atomic absorption spectrophotometer was used to determine the metal content. Chemical oxygen demand (COD) was measured according to standard methods [20].

### 2.3. Composition of the concrete mixes

The granules produced in series III were then used as coarse aggregate in concrete samples, prepared according to the UNI 11013 [21].

The characteristics of the granules used are described in Table 3. The reference aggregate (A3) was a calcareous one.

Table 3  
Selected properties of granules used for concrete sample preparation

| Aggregate | Binder           | F/FA ratio | Fluff (wt.%) |
|-----------|------------------|------------|--------------|
| A1        | CEM II-A/LL 42.5 | 3          | 50           |
| A2        | CEM II-A/LL 42.5 | 1          | 34           |

Table 4  
Mix-design of concrete samples

| Sample number                         | 1   | 2   | 3    |
|---------------------------------------|-----|-----|------|
| Aggregate                             | A1  | A2  | A3   |
| Cement (CEM II 42.5) (g/l)            | 350 | 350 | 350  |
| Water (g/l)                           | 175 | 175 | 175  |
| Aggregate (g/l)                       | 550 | 700 | 1325 |
| Siliceous sand (Torre del Lago) (g/l) | 500 | 500 | 500  |
| Water retention after 30 min (g)      | 113 | 51  | 6    |

The mix design of the samples is shown in Table 4. All the tests were performed in triplicate.

## 3. Experimental results

### 3.1. Product separation

From 10 t of the fluff produced in the plant the material passing the 4 mm sieve was separated: the remaining fraction was mainly constituted by foam and non-ferrous metals components. The passing fraction accounted for about the 28% of the total weight of the fluff and was mainly constituted by plastics, glass and inert materials. This fraction was sampled and subjected to analysis to evaluate, in particular, heavy metals concentration. Selected characteristics of this fraction are reported in Table 5.

### 3.2. Granulation tests

Figs. 2 and 3 show the experimental results of the granulation tests for series 1.

Results shows that increasing the water content with respect to the whole weight of the mix, also the average granules

Table 5  
Product characterization

| Parameter             | Value |
|-----------------------|-------|
| Residue at 105 °C (%) | 91.1  |
| Residue at 600 °C (%) | 39.4  |
| Copper (mg/kg)        | 3727  |
| Lead (mg/kg)          | 7420  |
| Chromium (mg/kg)      | <2    |
| Cadmium (mg/kg)       | 11    |
| Zinc (mg/kg)          | 450   |

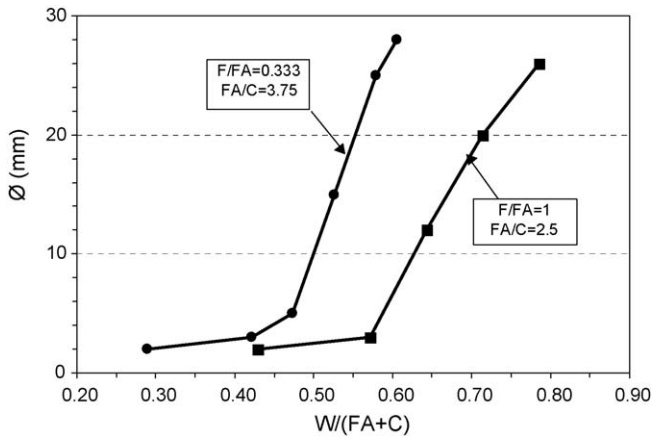


Fig. 2. Average granules diameter as a function of the water/binder ratio (series I).

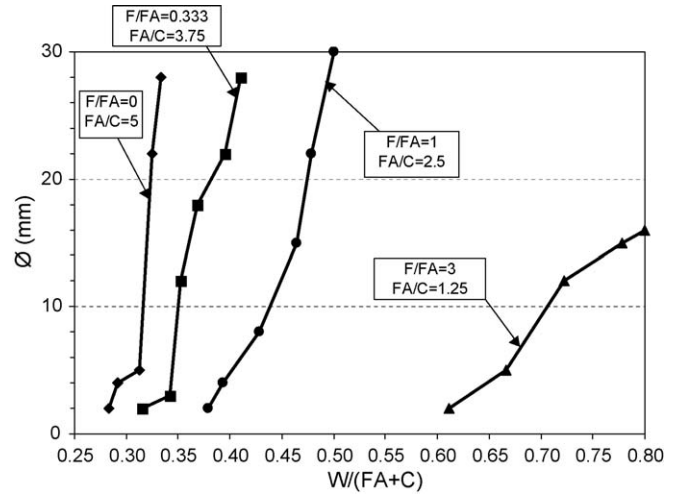


Fig. 4. Average granules diameter as a function of the water/binder ratio (series II).

diameter increased. A complete granulation was observed in the range of water addition between 22 and 48% of the solid weight. At a  $W/(F + FA + C)$  ratio of 0.48, a maximum diameter of 26–28 mm was achieved depending on the fluff content. A further increase in water content did not allow the granulation, and the formation of a sludge was observed. In addition, at any fixed water content, a higher fly ash content with respect to the binder content corresponded to a higher average granules diameter.

As regards the ratio between fluff and fly ash, Fig. 2 show that until  $F/FA = 1$ , an increase of the  $F/FA$  ratio resulted in a lower water consumption to produce granules of the same average diameter. This evidences a greater water request by the fly ash.

Figs. 4 and 5 show the results obtained in the series II.

It can be observed that the densifier plays a crucial role: it increases the slope of the curves that describe the influence of water content on granules average diameter. Consequently, in the presence of the densifier, a control of granules diameter can

be successfully performed. Figs. 4 and 5 show how this is not possible in the absence of the densifier. At the same time, in the absence of densifier, the granulation range is narrow.

As regards the  $F/FA$  ratio, the same consideration made for series I can be drawn: in addition, it can be noted that beyond  $F/FA = 1$  we observed an increase of water and a loss in granulation, that resulted in a lower maximum diameter achieved for the granules (at  $F/FA = 3$  a  $D_{max}$  of 16 mm was measured).

Figs. 6 and 7 show the results obtained in the series III, performed using cement as binder, and without any densifier addition.

Results obtained in this series show analogous behaviour to that obtained in series II, both in term of slope of the curves shown in Fig. 5 and in term of granules size.

In addition, it can be noted that the granulation range was the narrowest among the three series.

An image of selected granules produced is shown in Fig. 8.

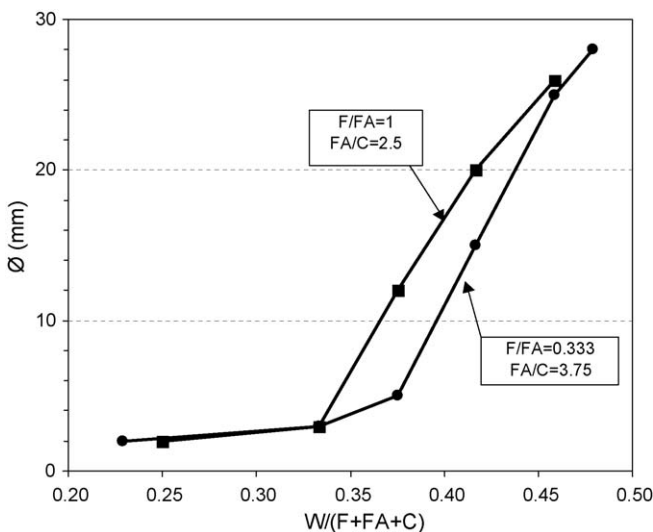


Fig. 3. Average granules diameter as a function of water content (series I).

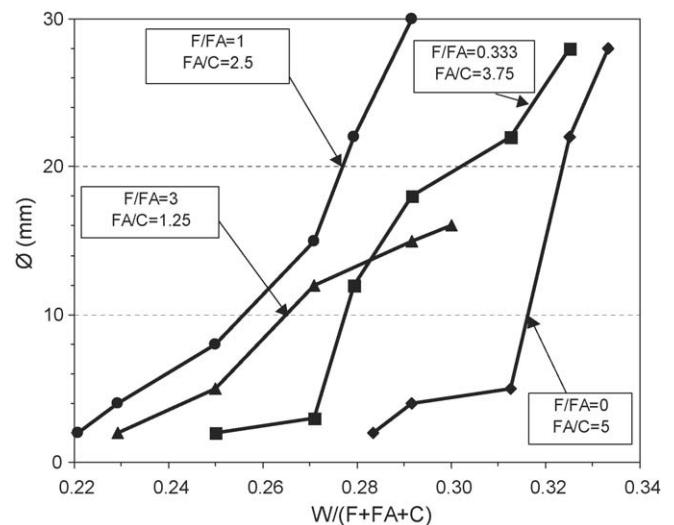


Fig. 5. Average granules diameter as a function of water content (series II).

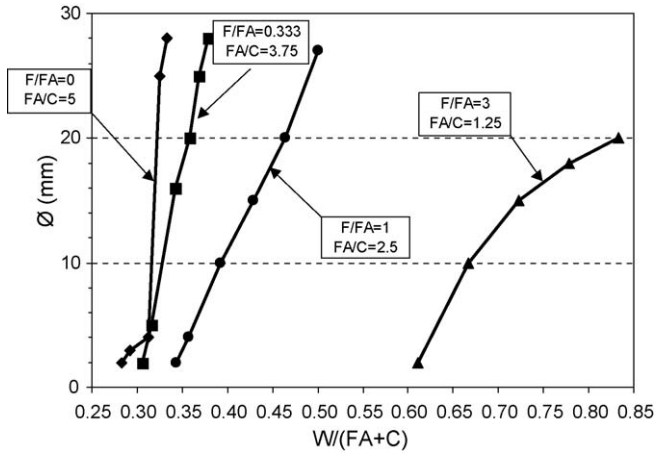


Fig. 6. Average granules diameter as a function of the water/binder ratio (series III).

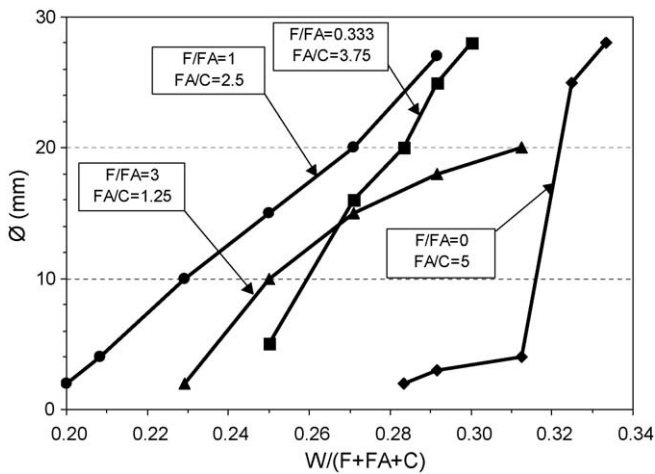


Fig. 7. Average granules diameter as a function of water content (series III).

Table 6  
Compressive strength tests on the produced granules

|            | Specific weight (kg/m <sup>3</sup> ) | Compressive strength (MPa) |
|------------|--------------------------------------|----------------------------|
| Series I   | 1250                                 | 1.0–1.2                    |
| Series II  | 1100                                 | 0.8–1.1                    |
| Series III | 1400                                 | 1.2–1.5                    |

3.3. Compressive strength tests

Table 6 shows the results of compressive strength tests performed on the granules.

Table 7 reports the preliminary results of compressive strength obtained for the sample prepared using selected granules as aggregate.

Table 7  
Compressive strength tests on concrete samples

| Sample | Specific weight of the aggregate (kg/m <sup>3</sup> ) | Water absorption [22] (%) | Specific weight of the sample (kg/m <sup>3</sup> ) | Compressive strength (MPa) |
|--------|---|---------------------------|--|----------------------------|
| 1      | 1100  | 21                        | 1700   | 7.84                       |
| 2      | 1400  | 7                         | 1800   | 14.28                      |
| 3      | –   | –                         | 2650   | 25.75                      |



Fig. 8. An image of selected granules produced along the experimentation showing diameters up to about 30 mm.

3.4. Leaching tests

Fig. 9 shows the results of the leaching tests performed on the granules: the only metal that may be released in the environment was zinc. Tests performed with the same procedure on the samples prepared using granules as aggregate show that no significant leaching of metals, ions and COD was detected.

4. Discussion

The results presented in the previous section showed that the fraction of fluff below 4 mm can be successfully immobilized in cementitious mixes by means of a granulation process. The granules may contain up to 50 wt.% of fluff, together with the necessary amount of binder (cement or lime), fly ash, and densifying agent. Depending upon the composition of the paste the specific weight of the granules varies between 1100 and 1400 kg/m<sup>3</sup>, resulting in a family of lightweight aggregates. The mechanical properties of these aggregates showed to depend both on water and fluff content. In fact, the results of tests performed on the granules showed that, as expected, higher compressive strengths and densities were achieved in the tests performed using cement

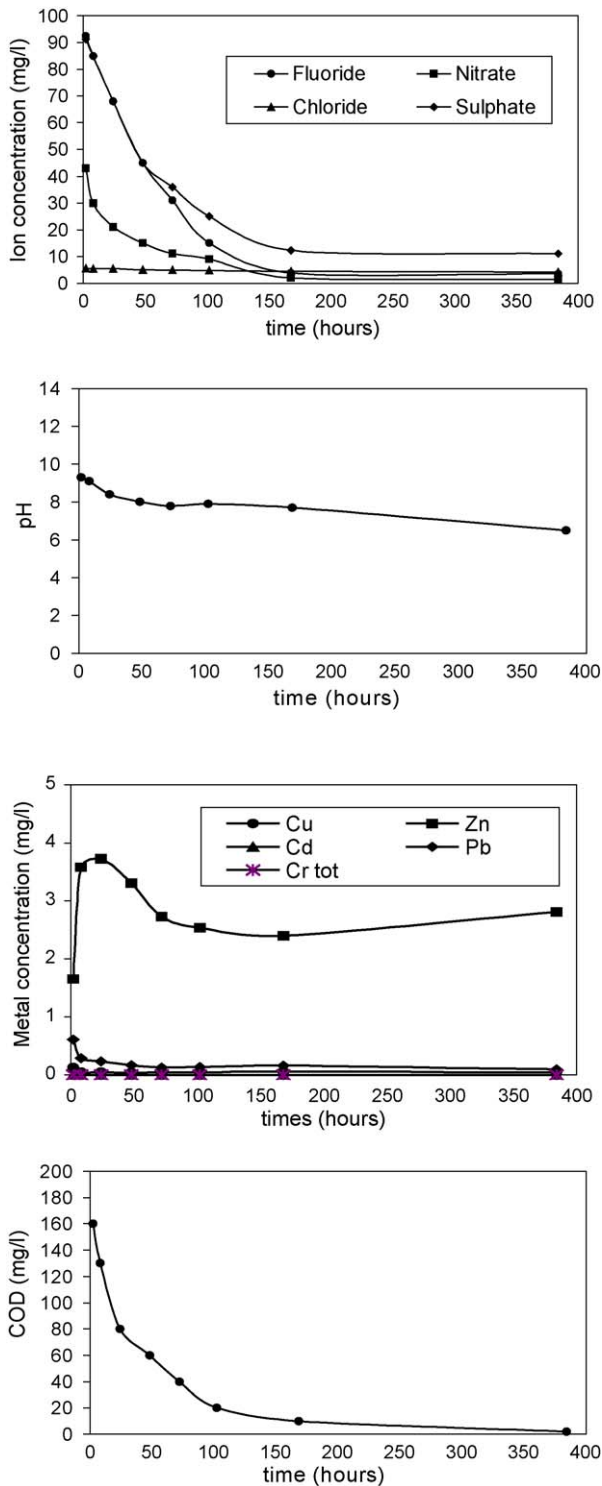


Fig. 9. Leaching tests performed on the produced granules.

as a binder. When lime was used as a binder, an increase in the mechanical properties of the granules was observed in the presence of the densifier.

The diameter of the granules produced was within a 5–30 mm range, depending upon water content. The densifier acts as a viscosizing agent, increasing the cohesion of the pellets during

the granulation process and reducing the increase of granules size at increasing water content.

Leaching tests performed directly on the granules showed a strong initial immobilising action, associated to the formation of the granules. The immobilization was effective for ions, organic compounds and heavy metals, with the only exception of zinc.

The two concrete sample prepared using the granules of aggregate, had a specific weight in the typical range of lightweight concrete [23]. In terms of strength, their average compressive strength was respectively about 30.5% and 55.5% of the calcareous reference sample. Such strength does not allow a structural use: according to the Italian Regulation UNI 7548 Part 2, the minimum allowed level of 15 MPa was not achieved. Additionally, it can be observed that the strengths achieved are quite low due to the high w/c ratio adopted in the mix-design (w/c = 0.67). For this reason, and also considering that the value reached when the fluff content was 34%, is very close to the strength limit, it can be reasonably assumed that a slight lowering in fluff content could easily satisfy the required strength condition.

The total absence of metals in the leachate from the tested concrete samples, confirms the further immobilizing action towards hazardous substances, as already observed in other immobilization studies [24,25].

The main advantage of the whole proposed process is the possibility of a complete reuse of the non-metallic fine fraction of automotive shredder residues.

This can be done successfully and directly without any preliminary further chemical treatment, to produce granules, which can be used as aggregate in lightweight concrete. As this fraction retains most of the pollutants of the fluff [25] the very strong immobilizing action that occurs, first during granules production and then in concrete preparation, ensures easily the complete inertization of any hazardous material. No additional thermal treatment is then required to produce aggregates compatible with Portland cement.

The separate coarse fraction, mainly constituted by plastic foam, and characterised by an high calorific heat, may be at the same time sent to incineration systems, depending upon its content in hazardous substances, thus avoiding landfill disposal and meeting the requirement of the more recent EU Directives.

## 5. Conclusions

In this paper, the non-metallic residue of an automotive shredder plant was granulated to produce aggregates for cementitious mixes.

The preliminary separation step produced a sieved fraction constituted mainly by inert non-metallic materials, which was characterised to investigate its metal content.

In the following granulation step, granules were produced by mixing this fraction with binding materials, fly ash and a densifier agent, in a rotating tank at room temperature.

Results show that the tested granulation technique can be successfully used to immobilize fluff from ASR and to regulate the size of the granules.

The size of the produced granules proved to be a function of water content: increasing the ratio between water and solids the diameter of the particles also increased.

Lime and cement, used as binders in the granulation phase, gave about the same results in term of granules size and mechanical strength. The use of a densifier allowed a larger range of granulation.

The maximum fluff (F)/fly ash (FA) ratio to allow granulation was pointed out at  $F/FA = 1$ : the obtained granules had 5–30 mm of diameter and up to  $1400 \text{ kg/m}^3$  of specific weight, while leaching tests showed that a good immobilization of metals and ions was achieved, except for Zn. This behaviour does not allow the use of the granules alone, but only as aggregate in a concrete mix.

Finally, preliminary concrete samples prepared using the produced granules as aggregate showed a specific weight up to  $1800 \text{ kg/m}^3$  and a compressive strength up to about 14 MPa, depending on the fluff content of the mixes, and on the nature of the binder and of other additives used.

Further efforts are however necessary, to achieve the optimal mix design of the granules and to investigate the possibility of their industrial production.

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