Production and characterization of aggregate from non

metallic Automotive Shredder Residues

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Abstract

In this paper the results of an experimentation on the granulation of non metallic automotive shredder residues to produce aggregates for cementitious or asphalt mixes are presented and discussed.

In a preliminary separation step a fraction containing mainly inert and non metallic materials was sieved to obtain the required grading and analyzed for the metal content. In the following granulation step, performed in a pilot scale granulator, the sieved fraction was mixed with binding materials, fly ash and a superplasticizer agent, to produce granules of up to 2000 kg/m³ specific weight. The size of the produced granules, between 2 and 40 mm, proved to be a function of water content: increasing the water/solids ratio, the diameter of the particles also increased.

The granules were then used as artificial lightweight aggregate for concrete mixes. Concrete samples showed a specific weight up to about 2000 kg/m³ and a compressive strength up to

about 30 MPa, depending on the fluff content of the mixes, and on the nature of the binder and of the other components used.

Leaching tests performed on the concrete samples showed that a good immobilization of metals and ions was achieved.

CE Database Subject Headings: automotive shredding residues; granulation; fly ash; artificial aggregates; lightweight concrete.

Introduction

Every year, in the UE about 12 M vehicles are shredded, and 8 or 9 Mtons/year of wastes are produced. This amount will probably increase in the future, as a consequence of the continuous expansion of the automotive industry. To minimize waste production, it is necessary to develop new strategies of management of the whole vehicles life cycle.

Right now two possible recovery pathways are outlined: the reuse of components, and the separation and reuse of the metal fraction (iron, aluminum, copper). 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, Automotive Shredder Residues) that account for about the 25% of the weight of a vehicle. Fluff mainly deals with rubber, glass, plastics, polyurethane foams, wood, textiles, paints, adhesives and non ferrous metals (Pèra et al., 2004). This residue is generally disposed in landfill as hazardous waste. As a consequence, until today, about 25% wt. of an end-life vehicle is landfilled, thus determining the contamination of soil and groundwater.

As a consequence of both the EU Directive about end life vehicles (2000/53/CE) together with the tightening of Environmental Regulation requirement for landfill disposal (EU Parliament, 1999; EU Parliament, 2000), and the increasing amount of automotive shredder

residues production, a new effort is necessary to find an alternative pathway for recycling and reusing the fluff fraction.

A promising solution could involve the concrete industry. There is an increasing difficulty in finding natural aggregates as a result of the years of use of the available rocks and minerals. Several studies have been performed to evaluate the possibility to reuse wastes in the production of aggregate for the concrete industry, possibly through an intermediate treatment step (Sagoe-Crentsil et al., 2001; Keyvani, 2002; Sani et al., 2005; Jantzen et al., 2001; Topcu et al., 2004).

Previous experimentations (Xu et al., 1995; Pera et al., 2004; Alunno Rossetti et al., 2006) 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 the work described in this paper, a selected fraction of the non metallic automobile shredder residues was immobilized in granules produced at room temperature in a pilot scale granulator, by mixing selected amount of fluff with a binder in the presence of additions and admixures. The granules were characterized and used as coarse aggregate in lightweight concrete mixes.

Materials and methods

Materials

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

In the granulation tests CEM I 32.5 R was used as binder. In order to increase the fluidity of the mixture and to reduce the water/cement /(w/c) ratio, a superplasticizer (ACE 363, Basf Construction Chemical Italia S.p.a.) was added to the mixing water (at 3% wt. with respect to cement content).

To enhance mechanical properties of the granules, the addition of fly ash produced in the thermoelectric plant of Brindisi, Italy, was also investigated. The composition and main characteristics of the fly ashes use are reported in Table 1.

Experimental procedure 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.

The proposed process deals with three steps: the selection of the product, the lab-scale granulation, the semi-pilot scale granulation.

In the first step plastics and foam were separated at the plant, by grinding the fluff produced and passing it through a 4 mm diameter mesh, while iron residues were separated with a magnetic system. Table 2 shows the characteristics of the selected product.

Pilot scale granulation tests were performed on the granulator device shown in figure 1 and designed to the purpose, basing on the results obtained in the previous tests performed in the lab scale granulator (Alunno Rossetti et al., 2006). Granulation tests were performed using in each test a total amount of mixture of 3-5 kg. The influence of the water content, fluff/binder ratio on the range of granulation and on granules diameter was evaluated.

The ratio between binder (C) and the sum of fly ash (FA) and fluff (F) in each test was 0.2. The weight ratio between fly ashes and fluff (FA/F) was selected at 0.83, 1 and 1.2, according to the results obtained in a previous work (Alunno Rossetti et al., 2006). Water

dosage was varied in a range from a minimum dosage necessary to obtain the granules, to a maximum dosage preventing the formation of a semifluid sludge in the granulator (water content granulation range). This value was considered as the optimal value for the granulation process.

After a 28 days period of curing at room temperature in a moisture saturated chamber, the granules were subjected to compressive strength tests, according to the UNI EN 13055-1:2003, and to leaching tests, according to the UNI EN 12457-4:2004. The 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.

The granules produced were then used as coarse aggregate in concrete samples, prepared according to standard methods (UNI 11013, 2002).

All the tests were performed in triplicate.

Results and discussion

Granules production

Pilot scale tests results confirmed the effectiveness of the granulation process. The size distribution obtained in the tests performed in the pilot scale granulator was strongly dependent upon the composition of the granules, as shown in Table 3. The size of the produced granules was a function of the water content. When the ratio between water and solids was increased, the diameter of the particles also increased. The tests when the average granules diameter values were higher were in fact the tests when the higher amount of water

was used (series D). In addition, a narrow diameter distribution interval was observed in the tests when a low water content was associated to the lower fly ash content.

The amount of granules produced in each test (distributed in four grading fractions: 20/40 mm, 12.5/20 mm, 4/12.5 mm and the fraction passing the 4 mm sieve), together with the granules resulting composition are summarized in table 4.

Depending upon the composition of the mixture, the specific weight of the granules varies between 1400 and 2000 kg/m³, originating a family of lightweight aggregates.

Mechanical properties of these aggregates, related to the specific weight, depend both on water content and fluff content: compressive strength values of the produced granules varied in the range between 0.8 and 1.5 MPa. As it would be expected, higher compressive strengths (1-1.5 MPa) and specific weights were achieved in the tests performed using the lower water/cement ratio.

ACV tests performed according to the BS 812-110 also showed that, depending upon the operating parameters in granules production, an increase of the fine fractions in the range between 25 and 35% was measured for the granules. The lower value was observed for the granules obtained in the series 3B. These results allow to consider the use of the granules, from a mechanical point of view, for road pavements.

Leaching tests

The results of leaching tests performed directly on the granules, are shown in Table 5, where the limit for inert and not hazardous wastes according to the EU Directive 2003/33/CE are also reported. Comparing these results with those of the raw material (Table 2) it can be assessed that a first strong immobilizing action with respect to heavy metals and organics was associated to the formation of the granules, with the only exception of a high COD

content measured in some tests. This exception could be due to both the presence of plastic residues in the sieved fraction, and to a slight contamination of the fluff by the fluids contained in vehicles tanks. This does not allow the use of the granules alone, but only as aggregate in a concrete mixture.

Concrete mixes characterization

In a first series of concrete samples production, the fractions 20/40 were used for the preparation of no fines concrete samples. The mix design parameter used are reported in Table 6, were the specific weight of the samples are also shown. A water/cement ratio of 0.5 was used in all the samples.

No fines concrete samples were characterized by means of leaching tests, according to Italian Regulation (Italian Environmental Regulation, 1998). Table 7 shows the results of cumulative leaching during the 16 days test. Results show that the granules still showed a significant release of organic matter, zinc and copper.

In a second series of tests, concrete samples were produced using respectively the 4/12.5 fraction, or the whole granules distribution below the 20 mm sieve, using a reference sand (Specific Weight=2690 kg/m³) as fine aggregate. The aggregate and the sand were proportioned, according to the Fuller distribution, obtaining a closer mix.

The composition of selected concrete mixes and concrete samples characterization in term of specific weight and compressive strength are reported in Table 8.

The results show that concrete samples prepared using the produced granules as coarse aggregates showed a specific weight up to 2000 kg/m³ and a compressive strength up to about 30 MPa, depending on the fluff content and composition of the mixes. The W/C ratio required to ensure the workability of each mix was also strongly dependent upon the total granules amount and size, together with the granules mechanical properties. As expected, in

the four samples prepared using only the granules fraction between 4 and 12.5 mm, very low W/C ratios were required, whilst the samples prepared also using the <4 mm fraction fractions, required an higher water amount to achieve the same workability. Due to the realization of a continuous grading of the aggregates, these last samples showed a higher compressive strength. In addition, since sample 3B was made of a higher amount of mortar (sand and cement) with respect to the other samples, it showed the higher specific weight and compressive strength. This sample was then subjected to leaching tests, according to the previously mentioned procedure (Italian Environmental Regulation, 1998). The results reported in Table 9 show that, as a consequence of the further immobilization action of the paste towards the granules 3B, the release of hazardous substances was strongly reduced: only small traces of zinc and copper and traces of organics were found in the leachate at the end of the leaching procedure, however in compliance with the limit imposed by Italian Environmental Regulation.

Conclusions

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

In a preliminary separation step a fraction made mainly of inert non metallic materials, in the form of a sand, with diameter less than 4 mm, was separated and characterised to determine the metal content.

In the granulation step, performed in a pilot scale granulator tank, granules were produced by mixing this fraction with cement, binding materials and a superplasticizer, at room temperature.

Results show that the granulation technique can be successfully used to immobilize fluff from automotive shredding residues and to adjust the size of the granules. The obtained granules had 2-40 mm of diameter and up to 2000 kg/m³ of specific weight: leaching tests showed that a good immobilization of metals and ions was achieved, except for organics. Preliminary results on concrete samples prepared using the produced granules as coarse aggregate showed a specific weight up to 2000 kg/m³ and a compressive strength up to about 30 MPa.

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| Total composition | | | | | | | | | |
|------------------------------------|-------|--|--|--|--|--|--|--|--|
| Component | % w/w | | | | | | | | |
| SiO_2 | 46.5 | | | | | | | | |
| Al_2O_3 | 24.4 | | | | | | | | |
| Fe_2O_3 | 10.1 | | | | | | | | |
| CaO | 7.0 | | | | | | | | |
| MgO | 1.1 | | | | | | | | |
| Na ₂ O+K ₂ O | 1.8 | | | | | | | | |
| SO_3 | 1.5 | | | | | | | | |
| L.O.I. (1100°C) | 5.2 | | | | | | | | |

Table 1 - Fly ash composition and characteristics

Specific surface (Blaine) 6800 cm²/g

| Parameter | Unit | 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 |

Table 2 – Product characterization

| | | Granules diameter (mm) | | | | | | | | | | | | |
|-----|----|------------------------|----|----|----|----|----|----|----|----|----|----|--|--|
| | 1A | 1B | 1C | 1D | 2A | 2B | 2C | 2D | 3A | 3B | 3C | 3D | | |
| Min | 8 | 2 | 2 | 22 | 6 | 2 | 2 | 15 | 2 | 2 | 2 | 10 | | |
| Max | 35 | 30 | 30 | 40 | 30 | 25 | 25 | 40 | 25 | 18 | 25 | 40 | | |

Table 3 – Granules range of diameters measured in the pilot scale tests

| Series | Fraction | Weight | W/C | FA/F | W/(C+FA) | Series | Fraction | Weight | W/C | FA/F | W/(C+FA) |
|--------|----------|------------|------|------|----------|--------|----------|--------|------|------|----------|
| | | (g) | | | | | | (g) | | | |
| 1A | 20/40 | 2506 | | | | 2C | 20/40 | 423 | | | |
| 1A | 12.5/20 | 568 | 1.08 | 1 | 0.41 | 2C | 12.5/20 | 475 | 0.92 | 0.83 | 0,31 |
| 1A | 4/12.5 | 83 | 1.00 | 1 | 0.11 | 2C | 4/12.5 | 1950 | 0.52 | 0.05 | 0,51 |
| 1A | <4 | 0 | | | | 2C | <4 | 190 | | | |
| 1B | 20/40 | 1013 | | | | 2D | 20/40 | 2054 | | | |
| 1B | 12.5/20 | 1888 | 1 | 1 | 0.375 | 2D | 12.5/20 | 1134 | 1.17 | 0.83 | 0,39 |
| 1B | 4/12.5 | 339 | | | 0.070 | 2D | 4/12.5 | 71 | 1117 | 0.02 | 0,69 |
| 1B | <4 | 128 | | | | 2D | <4 | 0 | | | |
| 1C | 20/40 | 731 | | | | 3A | 20/40 | 1059 | | | |
| 1C | 12.5/20 | 580 | 0.92 | 1 | 0.34 | 3A | 12.5/20 | 470 | 1.08 | 1.2 | 0,36 |
| 1C | 4/12.5 | 905 | | | | 3A | 4/12.5 | 1464 | | | - , |
| 1C | <4 | 672 | | | | 3A | <4 | 351 | | | |

| 1D | 20/40 | 3308 | | | | 3B | 20/40 | 0 | | | |
|----|---------|------|------|------|-------|----|---------|------|------|-----|------|
| 1D | 12.5/20 | 0.00 | | | 0.44 | 3B | 12.5/20 | 655 | | | 0.00 |
| 1D | 4/12.5 | 0.00 | 1.17 | 1 | 0.44 | 3B | 4/12.5 | 1880 | 1 | 1.2 | 0,33 |
| וו | 4/12.3 | 0.00 | | | | 30 | 4/12.3 | 1000 | | | |
| 1D | <4 | 0.00 | | | | 3B | <4 | 270 | | | |
| 2A | 20/40 | 1601 | | | | 3C | 20/40 | 485 | | | |
| 2A | 12.5/20 | 1289 | 1.08 | 0.83 | 0.41 | 3C | 12.5/20 | 678 | 0.92 | 1.2 | 0,31 |
| 2A | 4/12.5 | 133 | 1.06 | 0.63 | 0.41 | 3C | 4/12.5 | 898 | 0.92 | 1.2 | 0,31 |
| 2A | <4 | 0 | | | | 3C | <4 | 993 | | | |
| 2B | 20/40 | 1120 | | | | 3D | 20/40 | 2940 | | | |
| 2B | 12.5/20 | 852 | 1 | 0.83 | 0.375 | 3D | 12.5/20 | 402 | 1.17 | 1.2 | 0,39 |
| 2B | 4/12.5 | 1312 | 1 | 0.03 | 0.373 | 3D | 4/12.5 | 30 | 1.1/ | 1.4 | 0,39 |
| 2B | <4 | 162 | | | | 3D | <4 | 0 | | | |

Table 4 – Gradings produced and composition of the mixes

| pН | COD | Pb (mg/l) | Zn (mg/l) | Cd (mg/l) | Cu (mg/l) |
|-------|----------------------------------|----------------------------------|--|---|--|
| 10.86 | 40 | 0 | 0.35 | 0.05 | 0.22 |
| 10.53 | <20 | 0 | 0.04 | 0.05 | 0.22 |
| 10.65 | 197 | 0.21 | 0.53 | 0.05 | 0.22 |
| 10.79 | 473 | 0.06 | 0.04 | 0.05 | 0.23 |
| 10.86 | <20 | 0.05 | 2.62 | 0.05 | 0.16 |
| - | 50 | 0.05 | 0.4 | 0.004 | 0.2 |
| - | 80 | 1 | 5 | 0.1 | 5 |
| | 10.86 10.53 10.65 10.79 | 10.86 40 10.53 <20 | 10.86 40 0 10.53 <20 | 10.86 40 0 0.35 10.53 <20 | 10.86 40 0 0.35 0.05 10.53 <20 |

Table 5 – Results of leaching tests performed on selected granules (UNI EN 12457)

| Series | W/C ratio | W/(C+FA) | FA/F | MV |
|---------------------|-----------|----------|------|------|
| Concrete 1A (20/40) | 0.5 | 0.41 | 1 | 1.59 |
| Concrete 2A (20/40) | 0.5 | 0.36 | 1.2 | 1.66 |
| Concrete 1D (20/40) | 0.5 | 0.44 | 1 | 1.80 |
| Concrete 2D (20/40) | 0.5 | 0.39 | 0.83 | 1.52 |

Table 6 – Mix design of no fines concrete samples

| Test | pН | COD | Pb (mg/l) | Zn (mg/l) | Cd (mg/l) | Cu (mg/l) |
|-------------|---------|-----|-----------|-----------|-----------|-----------|
| | | | | | | |
| Concrete 1A | 11.2 | 20 | n.d | 0.5 | 0.001 | 0.02 |
| | | | | | | |
| Concrete 2A | 10.80 | <20 | n.d. | 6.2 | n.d. | 0.05 |
| | | | | | | |
| Concrete 1D | 11.15 | 40 | 0.01 | 3.5 | 0.001 | 0.05 |
| | | | | | | |
| Concrete 2D | 10.95 | 75 | 0.01 | 4.5 | 0.003 | 0.07 |
| | | | | | | |
| Limit | 5.512.0 | 30 | 0.05 | 3 | 0.005 | 0.05 |
| | | | | | | |

Table 7 – Results of leaching tests performed on no fines concrete samples

| Series | Cement | Aggregate | Sand | W/C | Rc ₂₈ | SW |
|-------------|--------|-----------|--------------|-------|------------------|----------------------|
| | (g) | (g) | (g) | ratio | (MPa) | (kg/m ³) |
| Concrete 2A | | | | | | |
| (4/12.5) | 392 | 581 | 855 | 0.26 | 18.06 | 1882.37 |
| Concrete 2B | | | | | | |
| (4/12.5) | 390.6 | 883.68 | 474.6 | 0.32 | 16.67 | 1800.64 |
| Concrete 2C | | | | | | |
| (4/12.5) | 390.6 | 797.27 | 619.8 | 0.30 | 27.27 | 1892.63 |
| Concrete 1B | | | | | | |
| (4/12.5) | 390.6 | 747 | 611.6 | 0.34 | 23.00 | 1911.87 |
| Concrete 3A | 390.6 | 858.88 | 409.6 | 0.38 | 25.77 | 1900.09 |
| Concrete 3C | 390.6 | 859.59 | 408.3 | 0.37 | 21.00 | 1819.53 |
| Concrete 1C | 390.6 | 868.52 | 392.2 | 0.38 | 24.21 | 1896.89 |
| Concrete 3B | 390.6 | 622.75 | 835.90 | 0.46 | 30.30 | 2013.88 |

Table 8 – Concrete composition and characterization

| Test | pН | COD | Pb (mg/l) | Zn (mg/l) | Cd (mg/l) | Cu (mg/l) |
|-------------|---------|-----|-----------|-----------|-----------|-----------|
| Concrete 3B | 11.35 | <20 | n.d | 0.1 | n.d. | 0.01 |
| Limit | 5.512.0 | 30 | 0.05 | 3 | 0.005 | 0.05 |

Table 9 – Concrete mix design and characterization