CHEMICAL ENGINEERING TRANSACTIONS
VOL. 43, 2015
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.aidic.it/cet
Chief Editors: Sauro Pierucci, Jiří J. Klemeš

ISBN 978-88-95608-34-1; **ISSN** 2283-9216
DOI: 10.3303/CET1543288

Artificial Aggregate From non Metallic Automotive Shredder Residue

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Until 2005 in the European Union (EU) approximately 12 M vehicles were yearly shredded, and 8 or 9 M t/ year of waste was produced. About 14 million tons of End of Life Vehicles (ELVs) are foreseen by 2015. This huge amount of waste must be treated and disposed of in a sustainable way. The most common treatment technologies, involve ELVs shredding to recover iron and steel (70%) and non ferrous metals (5%) from vehicles. The remaining fraction, called Automotive Shredder Residue (ASR), and representing about 25% wt. of each vehicle, is generally landfilled. For more than two thirds, this last residue deals with combustible materials (fibers, polyethylene etc..), suitable to be reused as a fuel, but a substantial amount of soil particles, metals, glasses and plastics residues are also present. Consequently, a new sustainable way to reuse ASR is to separate the organic from the inorganic fraction, and use them in combustion plants, gasification and in the cement industry, respectively. Regarding this second way of recovery, several studies have been already successfully performed with the aim of transforming ASR into aggregates for asphalt or cement mixes, by thermal treatment followed by chemical treatment, or by physical processes, such as granulation. In this work, a selected fraction of non metallic automobile shredder residue was immobilized in granules produced at room temperature in a pilot scale granulator. Granules were obtained by mixing selected amount of ASR with a binder (cement or lime) in the presence of additions (fly ash) and admixtures. The final aim of this work was to investigate the mechanical properties of concrete samples produced using the artificial aggregate obtained through different combinations of ASR, fly ash and binder. Additional freeze and thaw tests were finally performed to assess concrete durability along time.

1. Introduction

In general, a vehicle is made for approximately 75% of its weight from ferrous and non-ferrous, whereas the remaining 25% is made from organic materials (fine plastic) and silica (Fiore et al., 2012). Life end vehicles are generally subjected to several operations involving parts and components recovery before the final treatment which generally deals with shredding followed by mechanical and physical particles separation (Morselli et al., 2010).
Before shredding, fluids are removed, while tires, batteries, catalysts and all those components potentially toxic are recovered, to reduce the environmental impact of the subsequent phases. Even at this early stage it is possible to reuse selected components of the vehicles as spare parts.
The quantity and quality of the materials recovered at each step of the whole process depend on the type and characteristics of the vehicles. After milling, magnetic separation is commonly adopted to recover ferrous material, to send to metallurgical industries: the remaining non-ferrous materials and organic parts (glass, plastics, foams, rubbers, textiles) constitutes the so called “fluff”.
For more than two thirds of the overall amount, the fluff is constituted by fine combustible materials (fibres, polyethylene, etc.) with a calorific value higher than that obtained from biomass. However, this waste is highly contaminated by heavy metals such as lead and cadmium, and by the presence of oxides of copper and zinc, polychlorinated biphenyls (PCBs) and chlorofluorocarbons (CFCs). In addition, the fine fraction is the often contaminated by mineral oils and fluids. To reduce the environmental impact of such a complex waste, a new

Please cite this article as: Di Palma L., Medici F., Vilardi G., 2015, Artificial aggregate from non metallic automotive shredder residue, Chemical Engineering Transactions, 43, 1723-1728 DOI: 10.3303/CET1543288

disposal pathway has been proposed in the last decade by several authors, involving the separation of the organic from the inorganic fraction, and the use of the first one in combustion plants, gasification and the second one in the cement industry (Lam et al., 2010), after removal of big metallic particles (Pèra et al., 2004). Regarding the first way, the catalytic gasification of ASR for the generation of high-purity hydrogen has been recently proposed. ASR is partially oxidized and ultimately converted into hydrogen rich syngas (CO and H_2): the increase of the reaction temperature also favours the generation of hydrogen and the decomposition of the CO (Lin et al. 2010). For what concerns the second way excellent results have been already obtained by transforming the finest fraction of ASR (<4mm) into aggregates after a thermal treatment followed by a chemical treatment (Péra et al., 2004) or transforming it directly into concrete with the use of calcium sulfoaluminate cement or Portland cement (Alunno Rossetti et al., 2006).
In this work, a selected fraction of the nonmetallic automobile shredder residues was immobilized in granules produced at room temperature in a pilot scale granulator, by mixing selected amount of fluff with cement or lime and fly ash, in the presence of admixtures. The aim of this work was to study a possible improvement of the mechanical properties of the concrete as a result of a different combination of aggregates from fluff, fly ash and lime, also focusing on the durability and thus the maintenance over time of these characteristics.

2. Materials and Methods

2.1 Materials

Experimental tests were performed using the fluff produced in the automotive shredding plant “Italferro” located at Santa Palomba, Roma, Italy. In this plant, up to approximately 150 t of vehicles are shredded daily, and approximately 35 t of fluff are daily produced. The fluff (F) produced along three days of operation was sieved, and the fraction below 4 mm was collected and subjected to the experiments. In a first granulation test (S1) fluff is immobilized utilizing CEM I 32.5 R as a binder (C): to reduce the water/cement (w/c) ratio and to increase the fluidity of the mixture, a superplasticizer (ACE 363, BASF Construction Chemical Italia SPA) was added to the mixing water (at 3% by weight with respect to cement content). To improve the mechanical properties of the granules, fly ashes (FA) produced in the thermoelectric plant in Brindisi, Italy, were also used. Their composition and main characteristics are reported in Table 1.

Table 1: Fly ash composition and characteristics

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_2O+K_2O	1.8
SO_3	1.5
LOI (1100°C)	5.2
Specific surface (Blaine)	4800 cm^2/g

In a second series of granulation tests (S2), lime (L), used as a binder, and fly ash were used together with an accelerating agent (A). Two different amounts of agent dissolved in water were used: 0.14 and 0.08% w/w with respect to the lime. It was prepared by condensation of tetraethylenepentamine with formaldehyde (Medici et al., 2002). The reaction leads to the formation of polyaminoalkylolic oligomers, with free aminic and aminoalkylolic groups (Marruzzo et al., 2001).

2.2 Methods

2.2.1 Granulation

Pilot-scale granulation tests were performed on the granulator device shown in Fig. 1 and designed to the purpose, on the basis of the results from the previous tests performed at room temperature in a lab-scale granulator operating at 50 rpm (Di Palma et al., 2012).

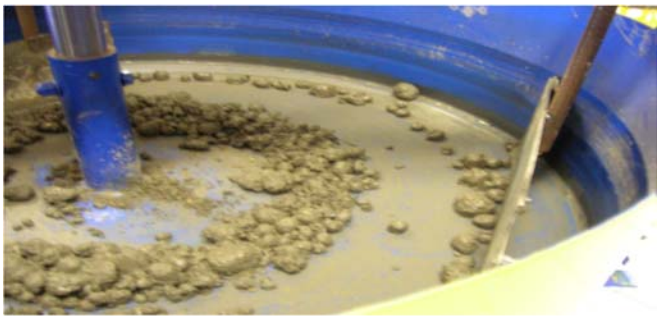

Figure 1: Pilot plant granulator

The granulation test deals with two steps: (1) the selection of the product and (2) the semipilot-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, and iron residues were separated with a magnetic system. Table 2 shows the characteristics of the selected product.

Table 2: Fluff fraction below 4 mm characteristics

Parameter	Unit	Value
Residue at 105°C	%	91.1
Residue at 600°C	%	39.4
Copper	mg/kg	3.7
Lead	mg/kg	7.4
Chromium	mg/kg	< 2
Cadmium	mg/kg	11.4
Zinc	mg/kg	450

Granulation tests were performed by using a total amount of mixture of 3-5 kg in each test. The ratio between binder and the sum of fly ash and fluff in each test was 0.2. The weight ratio between fly ash and fluff (FA/F) was selected at 0.83, 1, and 1.2, according to the results from a previous work (Alunno Rossetti et al. 2011). Water dosage was varied in a range from the minimum dosage necessary to obtain the granules to a maximum dosage, preventing the formation of a semifluid sludge in the granulator. This value was considered as the optimal value for the granulation process. After a 28 d period of curing at room temperature in a moisture saturated chamber (95 % relative humidity), the granules were subjected to compressive strength tests, according to the National Standard (UNI EN 2003), and to a leaching test, according to the UNI EN 10802: 2004. The specific weight of the granules was measured by hydrostatic weighing. The granulation tests produced 3 families of aggregates, whose compositions are reported in Table 3.

Table 3: Compositions of granules from S1

Families	W/C	FA/F	W/(C+FA)
SA1	0.92	0.83	0.31
SA2	0.92	1	0.34
SA3	0.92	1.2	0.31

For the granules production of the second type of binder (lime), 1000 g of ash were blended with 200 g of lime, utilizing the same binder/fly ash ratio of the previous test; the mix was then introduced into the previously described rotating drum. The amount of water or additive-solution sprayed during the first minutes of granulation was varied to achieve a mix with a good workability, subjectively evaluated. The formation and the accretion of granules took place within 2–5 min, keeping the rotating drum horizontal. When the granules reached the expected shape and dimension, they were collected and stored for 28 d of curing at room temperature and 95 % relative humidity. The specific weight of the granules was measured by hydrostatic weighing. From this second granulation series, three families of granules (named SB1, SB2, SB3) were obtained: their compositions are reported in Table 5, where the ratio between the accelerating agent (A) and lime is also shown. The compressive strength of granules was measured using a mechanical testing apparatus (J.J. Lloyd 20 T KN).

Table 4: Compositions of granules from S2

Families	L/FA	W/(L+FA)	A/L
SB1	0.2	0.7	-
SB2	0.2	0.7	0.14
SB3	0.2	0.7	0.08

2.2.2 Concrete samples preparation

Mixing the granules produced in the previous steps, according to the Fuller curve, six types of aggregate mixes were prepared, Ar90/20, Ar90/40, Ar60/40, Ar60/20, Ar90/60, Ar60/60, where the first number displays the ratio between S1 and S2, whereas the second one, indicates the ratio between (S1+S2) and gravel. These aggregates were then used to produce six concrete samples, as reported in Table 5. The tests were performed at a w/c ratio equal to 0.5, since according to previous works: this ratio ensured to the mix an adequate workability (Alunno Rossetti et al. 2006).

Table 5: Mix composition

Samples	Cement(g)	Gravel(g)	F(g)	CC(g)	Sand(g)	W/C ratio
Concrete Ar90/20	372	675	152	18	517	0.5
Concrete Ar90/40	372	511	306	34	522	0.5
Concrete Ar60/20	372	681.5	100	70	522	0.5
Concrete Ar60/40	372	517.5	207	138	528.5	0.5
Concrete Ar90/60	372	354.5	454	50.5	504.5	0.5
Concrete Ar60/60	372	360.5	308	205	513	0.5

Subsequently the w/c ratio of Ar90/20 and Ar60/40 was decreased to 0.44, according to mechanical properties and fluff contents. The freeze-thaw test was carried out only on these samples. Both aggregates and concrete samples were subjected to leaching test according to the UNI 10802:2004. The pH of the leachate was measured with a Crison 421 pH meter; a Philips PU 9200 atomic absorption spectrophotometer was used to determine the metal content, and an ionic chromatograph Dionex DX-120 was used to determine ionic species. The compressive strength was measured by a compressive strength tester KR150/M Tecnotest.

3. Results and Discussion

3.1 Granules Characterization

The granules from series S1, in which cement is utilized as binder, were characterized by a specific weight ranging between 1400 and 2000 kg/m^3, thus originating a family of lightweight aggregates. They were distributed in four grading fractions: 20/12.5 mm, 12.5/8 mm, 8/4 mm, 4/2 mm. Mechanical properties of these aggregates, related to the specific weight, were found to depend on water and fluff content; compressive strength of the produced granules were in the range between 0.8 and 1.5 MPa. The size distribution obtained in the tests performed in the pilot-scale granulator was strongly dependent on the composition of the granules and on the water content. When the ratio between water and solids was increased, the diameter of the particles also increased. The diameter of granules was in the range 2-25 mm.
As regards the granules obtained utilizing lime as a binder (series S2), they were characterized by a specific weight of the granules ranging between 1900 and 2300 kg/m^3 distributed in the same four grading fractions of aggregate from series S1. Compressive strength values were in the range between 0.2 and 1.2 MPa. These values were similar to those obtained in other studies: for instance, the results of compressive strength of lightweight aggregates obtained by thermal treatments of mixtures of fly-ash obtained from Serbian power plant ‘’Kolubara’’ and waste glass cover a range of 0.842-1.056 MPa (Zoric et al., 2012).
The results of leaching tests performed directly on the granules are showed in Table 6 where the limits for inert and not hazardous wastes, according to the EU Directive 2003/33/CE (UNI EN 2003), are also reported.
All the parameters are always below the limits, with the exception of COD. This can be attributed to the presence of plastic residues and organic traces (such as oil, foams and lubricants), which are absorbed onto the surfaces of waste particles. This exception excludes the possibility of the direct use of the granules and, as a consequence, a further immobilization step in concrete is required.

Table 6: Results of leaching tests

Test	pH	COD (mgO_2/L)	Pb(mg/L)	Zn(mg/L)	Cd(mg/L)	Cu(mg/L)
SA1	10.6	471	0.05	0.08	0.03	0.23
SA2	10.8	448	0.05	0.08	0.03	0.23
SA3	10.9	187	0.05	0.08	0.03	0.22
SB1	10.9	214	0.04	0.07	0.02	0.04
SB2	11.0	164	0.03	0.08	0.02	0.04
SB3	11.2	134	0.05	0.08	0.02	0.05
2003/33/CE inert		50	0.05	0.4	0.004	0.2
2003/33/CE not hazardous		80	1	5	0.1	5

3.2 Concrete Mixes Characterization

Mechanical and chemical properties of each concrete sample were investigated by leaching test, ionic chromatography, compressive strength test and freeze-thaw test.
Six concrete samples were prepared as indicated in Table 5, using the produced granules as coarse aggregates. Results of compression tests and values of specific weight are shown in the diagram below (Figure 2). The specific weight up to 2000 kg/m^3 and the compressive strength up to approximately 25 MPa, depending on the fluff content and composition of the mixes. Figure 2 displays on the horizontal axis the six series, which are organized following increasing amount of fluff. The average error for both measures is 5 %, considering the twenty investigated samples. It is possible to notice that, despite of the decrease of specific weight, corresponding to the higher amount of fluff, only a slight decrease of compressive strength was observed. By increasing fluff content of about four times the compressive strength decreased of around 5%. Table 7 shows the results of cumulative leaching during the 48 h test.

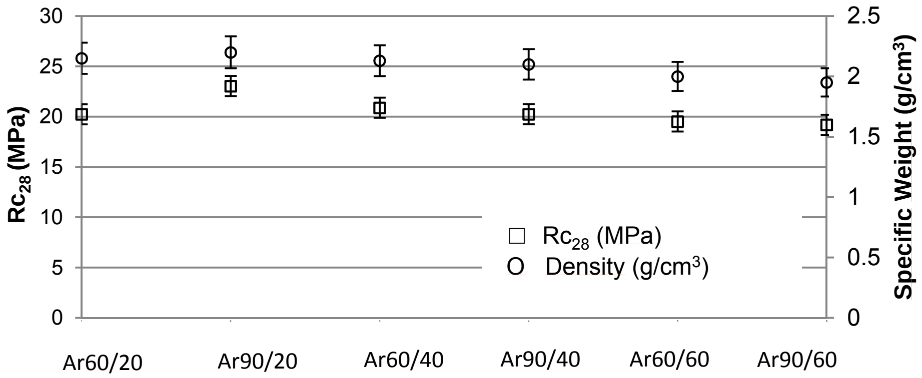

Figure 2: Specific weight and compressive strength

Comparing these results with those of the aggregate leaching test it is visible the widely decrease of the values of Pb, Cd and Cu. The most remarkable result is the significant reduction of COD content in the leachate, after the immobilization in concrete.

Table 7: Leaching test results on concrete samples

Test	pH	COD(mgO_2/L)	Pb(mg/L)	Zn(mg/L)	Cd(mg/L)	Cu(mg/L)
Concrete Ar90/20	11.5	<20	0.04	0.17	n.d.	0.02
Concrete Ar90/40	11.6	<20	0.03	0.11	n.d.	0.02
Concrete Ar60/20	11.4	<20	0.03	0.07	n.d.	0.01
Concrete Ar60/40	11.5	<20	0.04	0.09	n.d.	0.02
Concrete BAr90	11.5	<20	0.05	0.14	n.d.	0.02
Concrete BAr60	11.6	<20	0.03	0.12	n.d.	0.02
Limit	5.0-12.0	30	0.05	3.05	0.005	0.05

The freeze-thaw tests (ASTM C666) were carried out on the optimum mix of Ar90/20 and Ar60/40 in which the w/c ratio was decreased to 0.44. The results of the compression tests carried out after and before the freeze-thaw tests are reported in Table 8.

Table 8: Compressive strength before and after freeze – thaw tests

Sample	Specific weight (g/cm^3)	R_{c28} (MPa)	Specific weight (g/cm^3)	R_{c28} (MPa)
	Before freeze-thaw test		After freeze-thaw test	
Ar90/20	2.14	32.04	2.05	31.16
Ar60/40	2.12	26.78	2.04	26.05

According to the test results, concrete samples prepared by those aggregate mixes show a remarkable freeze-thaw resistance, which is fundamental to ensure the durability of the concrete itself.

4. Conclusion

A selected fraction of non-metallic automobile shredder residues was immobilizer in cement granules produced at room temperature in a pilot scale granulator and used as aggregate for concrete samples. Mechanical test results showed that these particular aggregates are suitable to be used in lightweight concrete. Increasing the amount of fluff immobilized in the granules, the specific weight reached 1.95-2.00 g/cm^3, despite of a negligible reduction of the compressive strength. In fact increasing the fluff amount of four times the decrease of compressive strength was less than 5%. Concrete samples also showed a noticeable freeze-thaw resistance, thus allowing the use of such a concrete in the North of Europe, where the lightweight concrete is the most utilized construction material.

References

Alunno Rossetti V., Di Palma L., Ferraro A., 2011, Production and characterization of aggregate from non metallic automotive shredder residues, Journal of Materials in Civil Engineering, ASCE, 23, 747-751, DOI: 10.1061/(ASCE)MT.1943-5533.0000189.

Alunno Rossetti V., Di Palma L., Medici F., 2006, Production of aggregate from non-metallic automotive shredder residues. Journal of Hazardous Materials, B137, 1089-1095.

Di Palma L., Mancini D., Medici F., 2012, Lab scale granulation tests of artificial aggregate production from marine sediments and industrial wastes, Chemical Engineering Transaction, 28, 199-204, DOI: 10.3303/CET1228034.

Fiore S., Ruffino B., Zanetti M.C., 2012, Automobile Shredder Residues in Italy: Characterization and valorization opportunities. Waste Management 32, 1548-1559.

Lam C.H.K., Barford J. P., McKay G., 2010, Utilization of incineration waste ash residues as Portland cement clinker, Chemical Engineering Transactions, 21, 757-762 DOI: 10.3303/CET1021127.

Lin K.S., Chowdhury S., Wang Z.P., 2010, Catalytic gasification of automotive shredder residues with hydrogen generation. Journal of Power Sources 195, 6016–6023.

Marruzzo G., Medici F., Panei L., Piga L., Rinaldi G., 2001, Characteristics and Properties of a Mixture Containing Fly Ash, Hydrated Lime, and an Organic Additive. Environmental Engineering Science, 18: 3, 159-165.

Medici F., Rinaldi G., 2002, Poly-Amino-Phenolic additives accelerating the carbonation of hydrated lime mortar. Environmental Engineering Science, 19: 4, 271-276.

Morselli L., Santini A., Passarini F., Vassura I., 2010, Automotive shredder residue (ASR) characterization for a valuable management. Waste Management 30, 2228-2234.

Péra J., Ambroise J., Chabannet M., 2004, Valorization of automotive shredder residue in building materials. Cement and Concrete Research, 34, 557-562.

Zoric D., Lazar D., Rudic O., Radeka M., Ranogajec J., Hiresenberger H., 2012, Thermal conductivity of lightweight aggregate based on coal fly ash, Journal of Thermal Analysis and Calorimetry, 110, 489-495.