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Is the assimilation to a solid recovered fuel a viable solution for automobile shredder residues' management?

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ABSTRACT

Directive 2000/53/EC and the European Circular Economy Package (2018) required the Member States to take all the necessary measures to reach the reuse-recycling goal of 85% for end-of-life vehicles (ELVs). In 2019, Europe achieved 89.6% of reuse-recycling, but most EC countries are still not completely compliant, Italy standing, for example, at only 84.2%. For this reason, actions are necessary to increase reuse-recycling for the waste generated in the operations of ELV shredding and separation, known as automobile shredded residues (ASRs). This study was aimed at assessing if the assimilation of ASRs to a solid recovered fuel (SRF) was a feasible solution. That would allow the waste to lose its status (end-of-waste, EoW), thus increasing the recycling rate. The assimilation of ASRs to SRFs requires the compliance with a series of parameters, namely net calorific value (NCV), content of chlorine (Cl), mercury (Hg) and selected heavy metals. The above-mentioned parameters were analyzed in the principal ASR fractions, namely textile, plastic and foam rubber, found in the samples collected during four sampling campaigns (2017–2021) performed at the same ELV treatment plant. Notwithstanding the great variability observed in the four samples, the results of the analyses revealed that the three fractions were compliant with NCV, Cl and Hg content. Conversely, the heavy metals' content was found a more critical parameter, in fact only the plastic fraction was suitable for SRF assimilation. Textiles presented criticality for the content of copper (Cu), nickel (Ni) and antimony (Sb). The heavy metals' contamination of foam rubber was found to be strongly related to particles' dimensions. A model which put particle size and metals' content into relationship was developed and validated. Removing particles of <40 mm significantly improved the quality of the material, however the content of Cu and Ni remained a critical issue for particles up to 200 mm. The SRF assimilation of the plastic fraction would increase the reuse-recycling rate of approx. 2.4–3.3%, thus allowing the achievement of the EC goals concerning the ELV management.

1. Introduction

The number of end-of-life vehicles (ELVs) worldwide is 50 million units per year, generating approx. 50 million tons of waste to be managed annually (Vijayan et al., 2022). ELVs are processed in authorized treatment facilities (ATFs), that is plants where, after the operations of depollution and dismantling, ELVs are shredded in order to liberate valuable fractions. Ferrous and non-ferrous metals are separated from the other materials, such as textile, foam rubber, plastic, rubber, glass, through a series of magnetic, eddy current, pneumatic and density separation processes (Cossu et al., 2014). Waste products generated from these processes are collectively named automobile shredder residue (ASR) and accounts for approx. 18–20% of an ELV (Santini et al., 2012; Cossu and Lai, 2015). ASR includes two main fractions, namely

light ASR and heavy ASR. Light ASR represents approx. 90% by weight (b.w.) of the overall ASR and it is generated in the correspondence of the main shredder, where an extractor fan aspirates dusts and the light pieces produced by the shredding operation (Fiore et al., 2012). According to previous studies, the main products found in light ASRs are textile, foam rubber and plastic (Karagoz et al., 2019). Conversely, rubber and large plastic scraps with a density of more than 1 kg/dm³ concentrate in heavy ASRs (Sakai et al., 2014). In recent years, automotive industries have incorporated lightweight materials, such as polymers and light metals, that reduce the weight of automobiles, but will enhance the generation of ASR in the future (Andersson et al., 2017; Khodier et al., 2018).

Directive 2000/53/EC, concerning ELV management, had fixed objectives of reuse-recycling at 85% of the vehicle's weight, and reuse-

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recovery at 95%, thus allowing only a 5% to be landfilled. According to the ELV Directive, reuse means “any operation by which components of ELVs are used for the same purpose for which they were conceived”. Recycling indicates “the reprocessing in a production process of the waste materials for the original purpose or for other purposes but excluding energy recovery”. Conversely, energy recovery means “the use of combustible waste as a means to generate energy through direct incineration with or without other waste but with recovery of the heat”. The recent European Circular Economy Package (2018) requires Member States to take all necessary measures to ensure that the storage and treatment of ELVs is carried out according to the waste hierarchy, without even changing the goals of reuse, recycling and recovery originally stated by Directive (2000)/53/EC.

According to Eurostat, in 2019, Europe achieved a reuse-recycling rate of approx. 90%, while the reuse-recovery rate was 95%. This result was achieved thanks to 11 EU Member States reporting reuse-recycling rates above 90%. However, the performances in recycling and recovery of ELVs in Italy and other EC countries was at low level, only approaching the reuse-recycling rate target fixed by the EC Directive (Italy, 84.2%). In Italy, the reuse-recovery rate was at the same value (84.2%), highlighting the lack of solutions/facilities for energy recovery.

Furthermore, the most recent data reporting the destination of the waste from ELV management in Italy, revealed that an amount equal to 87% of the light ASR is still disposed in landfills and only 12% and 1% are recovered through material or energy routes, respectively (ISPRA, 2022). This means that Italy currently does not meet the requirements of the European ELV Directive regarding landfill disposal. The reported low rates for ASR valorization are mainly due to several causes, such as (i) the high heterogeneity of the waste, (ii) the presence of fine shredding residues which contaminate the potentially valorizable/recoverable fractions (Gunaratne et al., 2020b) and (iii) the lack of plants ready to treat this waste (Bhari et al., 2021).

The net calorific value (NCV) of ASRs, in the range from 17 to 28 MJ/kg dry (Mancini et al., 2014), makes thermal processes an interesting option for ASR valorization through energy recovery, either directly, thus saturating the amount, equal to 10%, fixed by the EC Directive, or via the assimilation to a solid recovered fuel (SRF), thus increasing the reuse-recycling rate.

In the last few years there has been a growing interest in thermal treatments, such as pyrolysis and gasification. Such treatments should be capable of simultaneously producing energy and reducing the volume of the ASR residues to be disposed in a landfill. Most of the pyrolysis studies have been carried out at a lab scale, with the aim of investigating, for example, the quality of the gaseous emissions (Rey et al., 2016), the effect on the pyrolysis products' yield of the interactions among the polymeric components found in the ASRs (Yang and Chen, 2020), and the effect of a preceding process of torrefaction (He et al., 2023). Studies on gasification were aimed not only to find the best operating condition for syngas production, from either pure ASRs or mixed with other waste products (Lo et al., 2023), but also to investigate the characteristics of the obtained solid residue in the view of its valorization (Staničić et al., 2021; Yoo et al., 2022; Cho et al., 2023). However, pyrolysis and gasification technologies are deemed to be not completely mature for the treatment of ASRs at a full scale (Notarnicola et al., 2017; Vijayan et al., 2021). In fact, the presence of metals and the high heterogeneity of the waste made using ASRs to feed a pyrolysis/gasification reactor a difficult operation (Haydary et al., 2016; Ren et al., 2022).

Several other studies have also examined the feasibility of ASR combustion. For example, Lee (2007) characterized bottom and fly ashes coming from ASR incineration in order to use them as construction or lightweight aggregate material. The study of Lee (2007) could benefit of a commercial, full-scale plant located in Korea, a country in which the thermal valorization of ASR is a well-established process. Mancini et al. (2014) carried out ASR incineration tests at a full-scale tire incineration

plant with the aim of producing representative samples of bottom ashes and slags in order to study the feasibility of their recovery to obtain 85% recycling target. Hwang et al. (2008) tested some treatments, namely sieving, sink-float, aimed at improving the quality of ASR for fuel utilization. Edo et al. (2013) performed thermal decomposition tests with a lab-scale equipment of three light ASR fractions, namely <20 mm, 20–50 mm and 50–100 mm. The original sample contained a high amount of fine material (up to 50% b. w. < 20 mm). The low NCV and high emissions of polychlorinated dioxins and furans (PCDD/Fs), generated during combustion, made the fines fraction inappropriate for energy recovery. Conversely, the 50–100 mm fraction was found the most suitable to be used as a refuse derived fuel (RDF). However, the above-mentioned previous studies (Hwang et al., 2008; Edo et al., 2013) did not refer to an official definition of RDF, the quality of which was not assured and could vary from batch to batch. In order to make the operation of handling waste-derived fuel easier, a standardization was deemed necessary. The European Committee for Standardization (CEN), through CEN/TC 343 standard, introduced the definition of SRF, that is a fuel produced from non-hazardous waste in compliance with European standard EN 15359. The technical standard references which cover the operations of SRF production, sampling, analysis and characterization were elaborated by the CEN after solicitation of the European industry, primarily cement plants, interested in energy sources alternative to fossil fuels. However, the standardization at the European level did not concern the end-of-waste issue, thus remitting the legislation to the individual member states (Bessi et al., 2016). In Italy, Italian Ministerial Decree, DM 22/2013, established criteria for classifying SRFs no longer as a waste stream, but as a fuel, according to the end-of-waste approach. In order to be considered a fuel, the SRF must be produced according to the EN 15358:2011 quality system and is required to comply with the following issues: (i) being classified with a class code between 1 and 3 for NCV and Cl and between 1 and 2 for Hg (see Table S1); (ii) complying with the mandatory limits on micro-pollutants (Table 2 of Ministerial Decree n. 22/2013); (iii) the mechanical treatment plant needs to possess the REACH product registration. More recently, the Circolare of March 29, 2018 of the Italian Ministry for Environment and Land and Sea Protection (MATTM) included ASRs in the list of waste which can be assimilated to an SRF-fuel, provided the compliance with the criteria fixed by DM 22/2013.

This study had two main objectives. First, it examined the characteristics of the three main fractions, namely textile, plastic and foam rubber, extracted from the samples of ASRs collected at the same ATF during four sampling campaigns, carried out from 2017 to 2021. Textile, plastic and foam rubber fractions were analyzed in relation to the parameters, namely NCV, content of chlorine (Cl), mercury (Hg) and selected heavy metals, required for the assimilation of ASRs to an SRF. As mentioned before, previous studies have examined the feasibility of the thermal valorization of ASRs, but they did not focus on the

Table 1

NCV and ash content of the light ASR samples (I, II, III, IV – sampling campaigns).

	Plastic	Heavy textile	Foam rubber	<4 mm
NCV (MJ/kg)	36.6 ± 0.6	22.6 ± 0.5	28.0 ± 0.6	9.9 ± 1.3
DM 22/2013 class	1	2	1	5
Ashes I – II (%)	9.93 ± 0.35	21.46 ± 4.93	9.22 ± 2.78	57.34 ± 5.19
Ashes III (%)	4.80 ± 1.11	9.19 ± 0.31	5.14 ± 0.29	N.D.
Ashes IV (%)	N.D.	20.00 ± 4.25	0.72 ± 0.12 (a) 16.58 ± 1.08 (b)	N.D.

N.D. not determined; (a) sample extracted from the core of the particles, (b) sample extracted from the shell of the particles; I – II – III – IV: sampling campaigns.

Table 2

Metals' content of the samples of heavy textile, plastic and foam rubber and comparison with DM 22/2013 EoW limits.

	Heavy textile				Plastic				Foam rubber				DM 22/2013 EoW limits
	I Apr 2017	II Jul 2017	III May 2020	IV Sept 2021	I Apr 2017	II Jul 2017	III May 2020	IV Sept 2021	I Apr 2017	II Jul 2017	III May 2020	IV Sept 2021	
Sb	971	1352	83	101	17.7	14.2	10.7	17.0	86.1	28.3	4.48	7.95	50
As	14.7	<5	<5	<5	<5	<5	<5	<5	33.0	36.3	<5	<5	5
Cd	2.86	3.66	1.40	1.55	1.27	0.715	0.218	0.124	1.18	4.45	1.75	2.01	4
Cr	87.5	94.3	51.3	49.6	37.8	22.4	42.2	21.3	187	288	33.3	42.6	100
Co	9.42	10.9	6.76	6.73	12.6	8.27	4.55	12.8	9.79	27.8	3.54	6.95	18
Mn	135	164	93.9	129	18.1	12.7	11.0	8.89	122	358	58.6	88.6	250
Ni	83.8	103	43.1	217	10.6	7.22	7.65	5.67	201	1400	36.5	51.8	30
Pb	71.0	108	24.7	28.2	5.59	<3	7.62	5.04	36.7	158	29.8	29.3	240
Cu	13,400	3340	259	8130	16.7	11.8	151	78.2	801	1830	270	4290	500
Tl	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	5
V	3.89	5.44	<3	<3	<3	3.30	<3	<3	6.24	11.8	1.29	1.85	10

peculiarities required for the cessation of the waste status. If this recovery route, that is the assimilation of ASRs to an SRF, is proven to be feasible, it could contribute to increase the recycling rate of ELVs and consequently to be fully compliant with the targets set by the ELV Directive. Second, in the view of ASR thermal valorization, the study analyzed the composition of the combustion ashes obtained from each individual ASR fraction and compared it to that of wood pellets, a common combustible product utilized for energy generation in industrial and home furnaces. The findings of this study may be useful in future research targeted at identifying solutions for the reuse and/or disposal of ASR ashes.

2. Materials and methods

2.1. ASR and wood pellet: description and characterization analyses

The ASR samples used in this study were collected from the Centro Recuperi e Servizi ELV ATF located in Settimo Torinese (Metropolitan Turin Area, NW Italy). The ATF shreds and sorts both ELVs and metal scraps coming from old pieces of equipment (refrigerators, washing machines and other white goods) with an annual treatment capacity of 123,200 t (70 t/h). Ferrous and non-ferrous metals are separated through a series of operations, including sieving, magnetic, eddy current, pneumatic and densimetric separations, as comprehensively described in Ruffino et al. (2021).

Specifically, the ASR samples employed in this study were collected from the waste generated by the aspiration of the extractor fan located over the main hammer mill, during four sampling campaigns, carried out from 2017 to 2021, involving exclusively ELVs. The four sampling campaigns were carried out in April and July 2017, May 2020 and September 2021. The waste was sampled according to EN 15442:2011 standard, Solid recovered fuels – methods for sampling. The two samples collected in April and July 2017 were characterized and used already for the preliminary assessments reported in a previous work (Ruffino et al., 2021). The last sampling campaign (September 2021) was carried out during a full-scale shredding test which was aimed not only at producing a representative sample of light ASRs, but also to evaluate the re-use and recycling rate of the ATF according to EC Directive 2000/53. ELVs and the separated fractions were weighted before and after the shredding tests respectively. The separated fractions included the vehicle's parts removed in the dismantling phase (that is before the shredding operation) and the fractions separated in the sorting plant, that is (i) all the recoverable fractions, namely magnetic metals (proler), coarse (20–80 mm) and fine (<20 mm) zorba and the non-magnetic metals coming from the main eddy current (Titec) separator; and (ii) the waste products, namely the light and heavy ASR and dust. The sample collected in the September 2021 sampling campaign is shown in Fig. 1.

Light ASR samples underwent a product composition analysis aimed



Fig. 1. Sample of light ASR collected in the September 2021 sampling campaign.

at (i) the quantification of the principal materials which composed the samples and (ii) the separation of the fractions for further analyses.

The principal fractions of the ASR samples obtained from the product composition analysis, namely heavy textile, plastic and foam rubber (see Section 3.1), underwent a particle size analysis, by using a meter as a measuring instrument. All the fractions separated from the product composition analysis were ground in a Retsch SM 100 cutting mill to a final size of 1 mm. Ashes were obtained after a combustion at 950 °C of 5 ± 1 g of each milled fraction. The combustion was carried out in a muffle oven, with a maximum operating temperature of 1100 °C.

Two types of commercial wood pellets were also studied in order to compare the ashes produced by light ASR fractions and those produced by wood pellets. Wood pellet type n.1 was burned in the same muffle oven used to obtain ashes from the light ASR fractions. Wood pellet n.2 was burned in a domestic furnace and ashes were sampled from both the crucible (after an 8-h cycle of combustion, bottom ashes) and the flue (during the flue cleaning operation, after approx. a 5-month cycle of combustion with the same wood pellet material, fly ashes).

2.2. Analyses for the assimilation of ASR fractions to an SRF

On the fractions deemed of interest for the assimilation to an SRF, that is heavy textile, plastic and foam rubber, the parameters required by DM 22/2013 were determined, namely.

- NCV, according to EN 15400:2011 standard, Solid recovered fuels – determination of calorific value, with a bomb calorimeter after standardization with benzoic acid;
- Cl, according to EN 15408:2011 standard, Solid recovered fuels – methods for the determination of sulphur (S), chlorine (Cl), fluorine (F) and bromine (Br) content;
- Hg, according to EN 15411:2011 standard, Solid recovered fuels – methods for the determination of the content of trace elements, at an external laboratory specialized and accredited for waste analysis;
- The chemical parameters listed in Table 2 of DM 22/2013 (antimony (Sb), arsenic (As), cadmium (Cd), chromium (Cr), cobalt (Co), manganese (Mn), nickel (Ni), lead (Pb), copper (Cu), thallium (Tl) and vanadium (V)), according to EN 15411:2011 standard, through a two-stage acid digestion (Milestone Mega microwave oven, 0.25 g sample, first stage 3 mL H₂SO₄, second stage 3 mL HNO₃) and subsequent ICP-OES analysis (Perkin Elmer, Optima 2000 DV);

The above determinations were made on the ground samples as specified in Section 2.1.

In the case of a three-dimensional product such as foam rubber, the measured metal concentration appeared to be related to the particle size distribution (see Section 3.2). Consequently, in order to find a relationship between metals' content and particle size, it was assumed that the overall amount of metals found in the samples came from a combination of a clean inner core and a dirty few-mm thick shell. The pieces of foam rubber were modelled as cubes with a 3-mm thick shell and (n-6) mm inner core ("core-shell" model). Shell and cores, coming from the foam rubber pieces sampled in the fourth sampling campaign (September 2021) were ground and analyzed separately, in order to assess the metals' concentration of the clean and dirty zones. For metals' determination, the same method of above was used.

3. Results and discussion

3.1. Characterization of the ASR samples

The mass balance accounting the fractions separated during the shredding test performed in September 2021 revealed that the reuse-recycling rate of ELVs at the ATF was equal to 82.2%. That amount included the vehicle's parts removed before the shredding operation (namely air bags, bumpers, tires, battery, engine, gas tank, catalyzers, wheel cups, starter, which accounted for 16.6% of the ELV weight) and the fractions separated in the sorting plant, which were magnetic metals (proler, 61.2% b. w.); coarse (20–80 mm) zorba (3.1% b. w.); fine (<20 mm) zorba (0.7% b. w.) and non-magnetic metals from the main eddy current (Titec) separator (0.5% b. w.). The fractions excluded from recycling were light ASR, heavy ASR and dust which accounted for 16.3%, 1.1% and 0.5% b. w., respectively. It was apparent that the achieved re-use and recycling rate was lower than the goal fixed by the EC Directive (85%), although the ATF had worked at its highest efficiency in performing the operations of dismantling and sorting during the shredding test. Consequently, actions are needed to increase re-use and recycling, for example through improved operations of metal separation from the light ASRs and/or assimilation of fractions with high NCV to an SRF.

The results of the product composition analysis carried out on the four samples of light ASRs are shown in Fig. 2.

The samples collected in the four sampling campaigns showed a great variability of composition, but it can be seen that, in almost all samples of the light ASRs, four fractions, namely heavy textile, plastic, foam rubber and particles with sizes of less than 4 mm, were predominant. The amounts of heavy textile, plastic and foam rubber were in the order of 22–25%, 15–20% and 10–12% b. w., respectively. These figures were partially in line with the values reported in recent studies carried out in other parts of the world. Yang and Chen (2020), for example, found a similar amount of textiles, but a higher amount of plastic

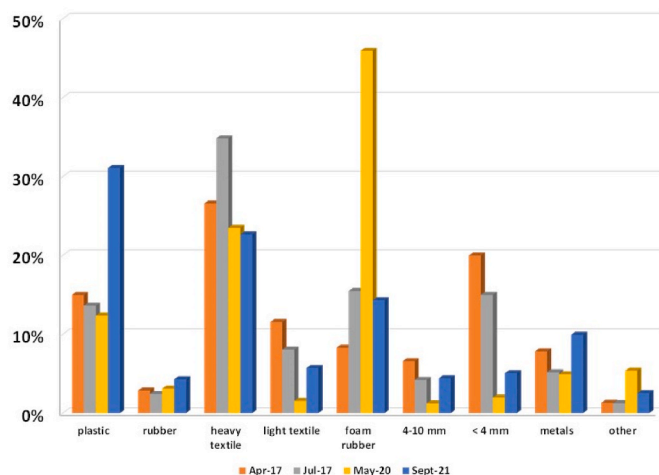


Fig. 2. Results of the product composition analysis of the four samples of light ASRs.

(approx. 40%) and minor quantities of foam rubber (approx. 2%), in a ASR sample obtained at an ATF in China. The abundance of particles with sizes of less than 4 mm differed between the first and the last couple of samples by a large extent, being between 15 and 20% b. w. in the 2017 samples and of only a few percentage units in the samples collected in 2020–2021. Amounts of foam rubber and plastics, higher than the average, were present in the third and fourth sample, respectively. Rubber and particles with sizes 4–10 mm were found to be minor fractions, with an abundance lower than 5% b. w. The amount of residual metals in the light ASRs ranged between 5% and 10% b. w., thus probably making processes of enhanced separation worthy to be performed, in order to improve metals' recovery efficiency and, consequently, the overall re-use and recycling rate.

3.2. Tests for SRF assimilation

The SRF classification consists of the assignment of a class code, from 1 to 5 (see Table S1), according to EN 15359 standard, Solid recovered fuels – specifications and classes, and DM 22/2013, on the basis of statistical evaluations performed on a population of ten (10) consecutive analyses for each of the three classification parameters: NCV (related to economic value), Cl content (related to technical issues) and Hg content (related to environmental issues). Furthermore, the waste must comply with two physical parameters, namely ashes and moisture, and a number of chemical parameters (As, Cd, Cr, Co, Cu, Mn, Ni, Pb, Sb, Tl, V) listed in Table 2 of DM 22/2013 and foreseen by EN 15359 standard.

The fractions of heavy textile, plastic and foam rubber could be considered of interest for assimilation to an SRF, because of (i) their abundance into light ASRs and (ii) NCV, which is one of the crucial input data for the design of a thermal process (Haydary and Susa, 2015). The above-mentioned fractions accounted for approx. 50% b. w. of the overall light ASR samples. Furthermore, as it can be seen from Table 1, NCVs of plastic and foam rubber placed into class 1, and heavy textile's NCV into class 2, according to standard EN 15359 and DM 22/2013. The fraction with dimension of less than 4 mm had a NCV too low to be considered for SRF assimilation, in fact its value placed into class 5. Other valorization routes, such as utilization as substitute raw material in mortar or concrete, or metal extraction through pyro- (Jagodźnińska et al., 2021) or hydrometallurgical processes (Gao et al., 2021), are currently under evaluation for shredder fine residues (Gunaratne et al., 2020a).

Consequently, on the grounds of the sole NCV parameter, heavy textile, plastic and foam rubber were deemed good candidates for the assimilation to an SRF. However, other than NCVs, classification of the

ASR fractions to an SRF requires the compliance with Cl, Hg and metals content.

Repeated analyses of Cl and Hg on the three fractions extracted from the four ASR samples demonstrated that the three materials had the quality requirements required by the standard to be placed into class 1. EN 15359 standard does not fix specific requirements for ashes and moisture; suitable values of the two parameters depend on agreements between SRF producer and utilizer. For all the analyzed ASR samples, moisture was in the order of 2% b. w. and the values of ashes are listed in Table 1. On the basis of a preliminary, rough observation of the light ASR samples, it was noted that the residual quantity of ashes was related with the size distribution of the particles of the analyzed fractions (see Fig. 3). Fractions with smaller particles, like those coming from the September 2021 sample, had, on average, higher amounts of ashes.

Table 2 reports the median concentration value of the 11 metals listed in EN 15359 standard for the fractions of heavy textiles, plastic and foam rubber extracted from the four light ASR samples. It can be seen from the results of Table 2 that the concentrations of As, Cd, Cr, Co, Tl and V found in the heavy textile samples had almost constant values along the four sampling campaigns and, generally, were below the limits fixed by DM 22/2013. A higher fluctuation was observed for Cr and Pb, however the concentration values of the two metals were below the limits. Conversely, because of the concentrations found for Sb, Ni and Cu, the assimilation of heavy textiles to an SRF was not allowed. Amounts of Ni and Cu were of the same order of magnitude of those reported in the study of Mancini and coauthors (2014), even if the values of that study were referred to the overall samples of light ASRs.

The values of Table 2 show that amounts of Ni and Cu above the limits fixed by EN 15359 and DM 22/2013 were found also in some of the samples of foam rubber. Conversely, the criticality on Sb seemed to be a characteristic of heavy textile, which exceeded the limits in all samples, because antimony oxide (Sb_2O_3) is used as a catalyst in the processing of base materials for the production of polyester fibers and filament yarns (Liu et al., 2021) and as a fire-retardant additive in synthetic textiles (Babushok et al., 2017). Textiles are porous materials, consequently contaminants, especially metals, which concentrate in the fine dust resulting from the operations of primary and secondary shredding, can penetrate into such a porosity and be entrapped into it. That intrinsic characteristic of textiles makes difficult to carry out operations aimed at de-polluting such materials and, consequently, make them compliant with the requirements fixed by DM 22/2013.

As it can be seen from Table 2, promising results were obtained for plastics. The concentration of all metals found in the samples were below, in the case of most metals well below, the limits fixed by DM 22/

2013, thus allowing the assimilation of the plastic fractions to an SRF. Unlike textile, plastic pieces had a limited or no porosity, consequently the polluted fine dust could not penetrate into them and the amount of retained contaminants was minor. As shown in Fig. 2, on the basis of the data collected in the four sampling campaigns carried out between 2017 and 2021, the average amount of plastics into ASRs was in the order of 15–20% b. w. The separation of plastic pieces from the light ASRs and assimilation to an SRF could increase the recovery rate by approx. 2.4–3.3%, thus rising it from 82.2% to 84.6–85.4%.

The values of Table 2, for what concerns the metals' content found in the foam rubber samples, highlight a quite high variability among the four samples. On average, it can be seen that the foam rubber fractions coming from the samples collected in 2017 had a worse quality if compared to that of the two more recent samples. The most critical metals for the assimilation of foam rubber to an SRF appeared Cu and Ni, as already observed for heavy textile, but also As, Cr, Co, Mn and Sb required a careful and more precise evaluation.

On the basis of the results reported in Table 2, two observations can be made. On the one hand, as it can be seen from Fig. 2, a clear relationship could be identified between metals' content and amount of fine particles (<4 mm). In fact, the samples collected in 2017, which were characterized by a higher metal content, also contained a higher amount of the <4 mm fraction, with respect to the samples which dated back to 2020 and 2021. The amount of the <4 mm fraction was equal to approx. 20% and 15% b. w. in the two samples collected in 2017 (April and July, respectively), and it was only approx. 2% and 5% b. w. in the samples collected in May 2020 and September 2021. Shredder fine residues covered and made the surfaces of the ASR pieces dirty. This may determine contamination of the material and prevent its assimilation into the SRF. On the other hand, as observed for ashes, also the concentration of metals seemed to be related to the size distribution of the foam rubber particles. This issue was deemed worthy of a further investigation; a correlation was then searched for between the size of the foam rubber particles and the concentration of metals, and the way that affected the possibility of assimilating foam rubber to an SRF.

The particle size distribution of the two foam rubber samples (05/20 and 09/21) is shown in Fig. 3. Precise data concerning the particle size distribution of the 2017 foam rubber fraction were not available, their D50 was approx. 50 mm. The D50 for the two foam rubber fractions collected in 2020 and 2021 was of 150 mm and 120 mm respectively, however the 09/21 sample had a particle size distribution narrower than that of the 05/20 sample. An amount equal to 50% b. w. of the 05/20 sample had dimensions ranging from 150 to 330 mm; conversely, 50% of the sample collected in September 2021 had dimensions between 120 and 140 mm, and no particles with sizes larger than 140 mm.

The relationship between metals' content and particle size was searched for as described in Section 2.2, with the pieces of foam rubber modelled as cubes with a 3-mm thick shell and (n-6) mm inner core. Average metals' values of the clean and dirty zones are reported in Table S2. Table S2 also reports calculated metals' content for pieces with sizes of 40, 80, 120, 160 and 200 mm. The results of Table S2 show that the inner core of the foam rubber particles contained very little amount of metals, in some cases below the detection limits of the ICP-OES. The analyses of the shells revealed that the content of As, Pb, Sb, Tl of foam rubber was compliant with the EoW requirement for SRF assimilation even in the worst case, that is for whichever size of the particle. In fact, the contents of As, Pb, Sb, Tl found in the 3-mm thick particles' shells were lower than DM 22/2013 limits. Fig. 4 reports the ratio between the metal contents found in the shells and the limits of DM 22/2013, only for the metals the content of which exceeded the limits of DM 22/2013. The ratio was calculated with the average value and the maximum value, that is average plus standard deviation. It can be seen from Fig. 4 that the content of Cd, Cr, Co, Mn and V found in the shells exceeded DM 22/2013 limits by 1.5–3 times, suggesting that foam rubber could be assimilated to an SRF after elimination of the particles with sizes below a certain threshold value. In fact, the above-mentioned "core-shell" model

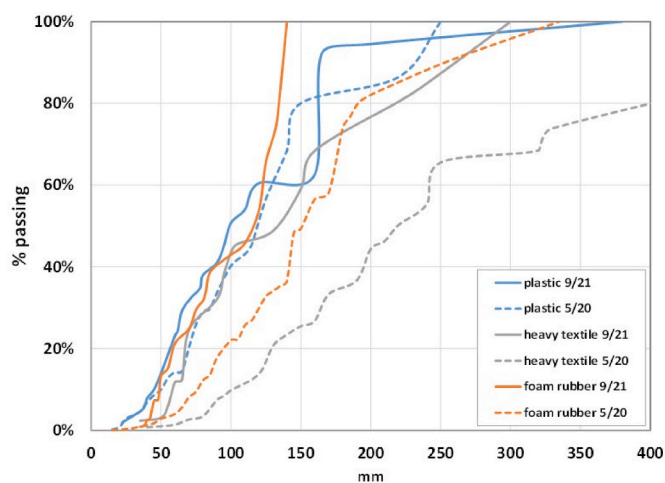


Fig. 3. Results of the particle size analysis of the fractions of plastic, heavy textile and foam rubber separated from the samples collected in 2020 and 2021. The results for the 2017 samples are not available.

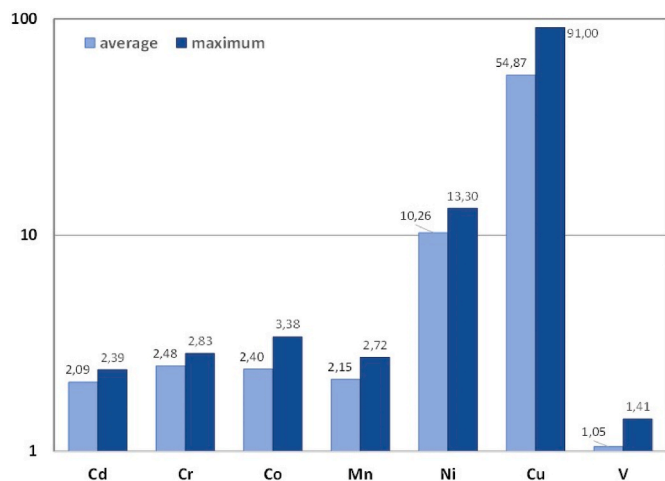


Fig. 4. Ratio between the metal content found in the foam rubber particle shells and the limits of DM 22/2013 (only for the metals the content of which exceeded the limits of DM 22/2013). The comparison was made with the average value (light blue bars) and the maximum value (dark blue bars), that is average plus standard deviation.

is based on the dilution operated by the clean core on the overall foam rubber particle contaminated by the exterior shell. On the basis of the results reported in Table S2 and Fig. 4, the situation for Cu and Ni appeared quite difficult, because the two metals exceeded the limits fixed by DM 22/2013 by approx. 10 times and over 50 times, respectively. The results coming from the application of the “core-shell” model to the foam rubber particles suggests that, in the presence of limited amounts of the <4 mm fraction, around 5%, foam rubber particles with sizes of more than 40 mm had metals’ contents which were compliant with the EoW requirements, with the exception of Cu and Ni. The amount of Ni observed in the shells of foam rubber particles would require pieces with sizes of more than 200 mm to be compliant with the EoW requirements. Conversely, in no cases, the amount of Cu found in the foam rubber separated from light ASRs allowed the assimilation of such a fraction to an SRF.

Fig. 5 shows the trend of metal concentration, as a function of the foam rubber particle sizes, predicted by the model, together with the values found in the samples collected in the sampling campaigns. The straight lines represent the threshold values fixed by DM 22/2013 and EN 15359 standard. The content of Co, Cr, Cu, Mn and Ni found in the foam rubber samples was related to the D50 of the particle size distribution, that is D50 = 50 mm, 150 mm and 120 mm for the samples collected in 2017, 2020 and 2021, respectively. It can be seen that the results predicted by the model were in good agreement with the metal content observed in the analyzed foam rubber samples.

3.3. Heavy metals evaluation in ASR thermal residues

The results shown in Section 3.2 demonstrated that, among the main fractions found in the light ASRs, the sole plastic could be assimilated to an SRF according to EN 15359 standard and DM 22/2013. However, the NCVs of heavy textile and foam rubber made these fractions of interest for energy valorization, in order to saturate the amount of 10% of thermal valorization admitted by Directive (2000)/53/EC, achieve the 95% goal of re-use and recovery, and reduce the disposal in landfills. For this reason, with the aim of providing useful information for the thermal valorization of ASRs, the composition of the ashes generated from the combustion of each single fraction separated from the light ASRs was determined and compared with that of (i) the ashes coming from the thermal valorization plant of the MSW fraction that remains after separate collection in the city of Turin (Italy) and (ii) the ashes generated from the combustion of wood pellets. None of the previous works

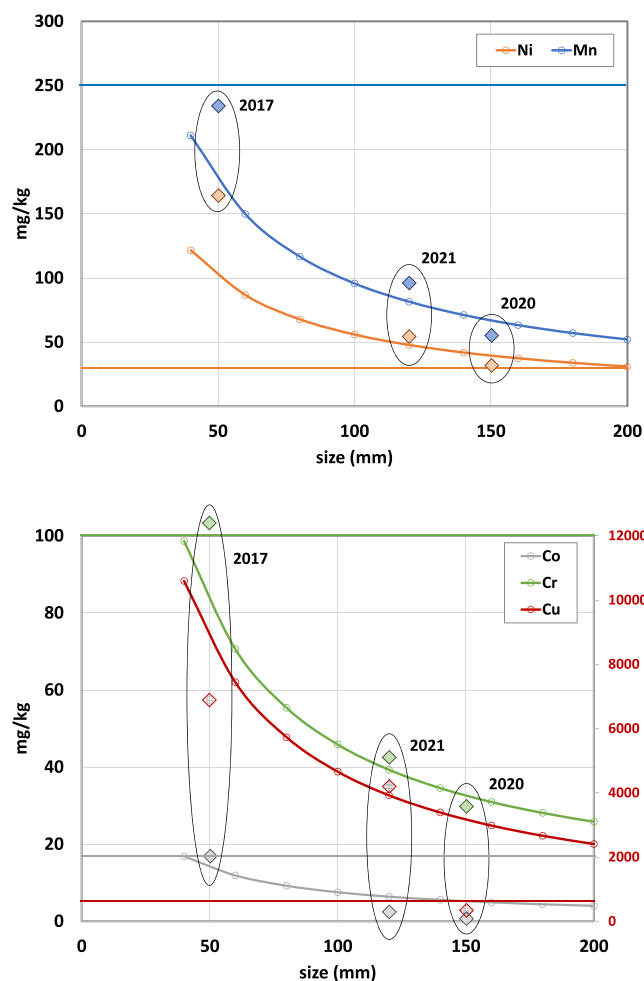


Fig. 5. Trend of metals’ concentration predicted by the model, together with the values found in the samples and threshold values fixed by DM 22/2013 and EN 15359 (straight lines).

on ASR thermal valorization provided the characterization of each single fraction of ashes. Information is available on the quality of ashes generated from the combustion of entire samples of ASRs (Lee, 2007; Mancini et al., 2014; Lee et al., 2015).

The results of the ashes characterization (Cd, Co, Cr, Cu, Mn, Ni, Pb, Zn) are reported in Fig. 6. The characterization of the ashes coming from the thermal valorization plant was provided from the plant’s managers. The plant located in the Metropolitan Turin Area has an authorized capacity of 421,000 t/y for waste with an average NCV in the order of 11 MJ/kg (Crivello, 2015). The metals’ content values in the wood pellet ashes were an average of three replicates. The metals’ content values in the ashes from the ASR fractions were an average of twelve replicates (three replicates for each fraction collected in each of the four sampling campaigns).

It can be seen from Fig. 6 that Cu and Zn were the metals with the highest concentrations in the ASR ashes. A high fluctuation in the Cu concentrations among the different ASR fractions could be observed. The highest amount, in the order of 7%, was found in the light textile (LT), that is the fluffy material generated from the tearing of heavy textile. Such a material has a great capacity of retaining metallic wires, in fact, it showed the highest concentration of metals, among the ASR fractions, also for Ni and Pb. The combustion of all the ASR fractions, with the sole exception of plastic, generated ashes with a Cu content well above (10–70 times) MSW ashes. However, such a high Cu concentration makes those ashes valuable for metal recovery, as an alternative

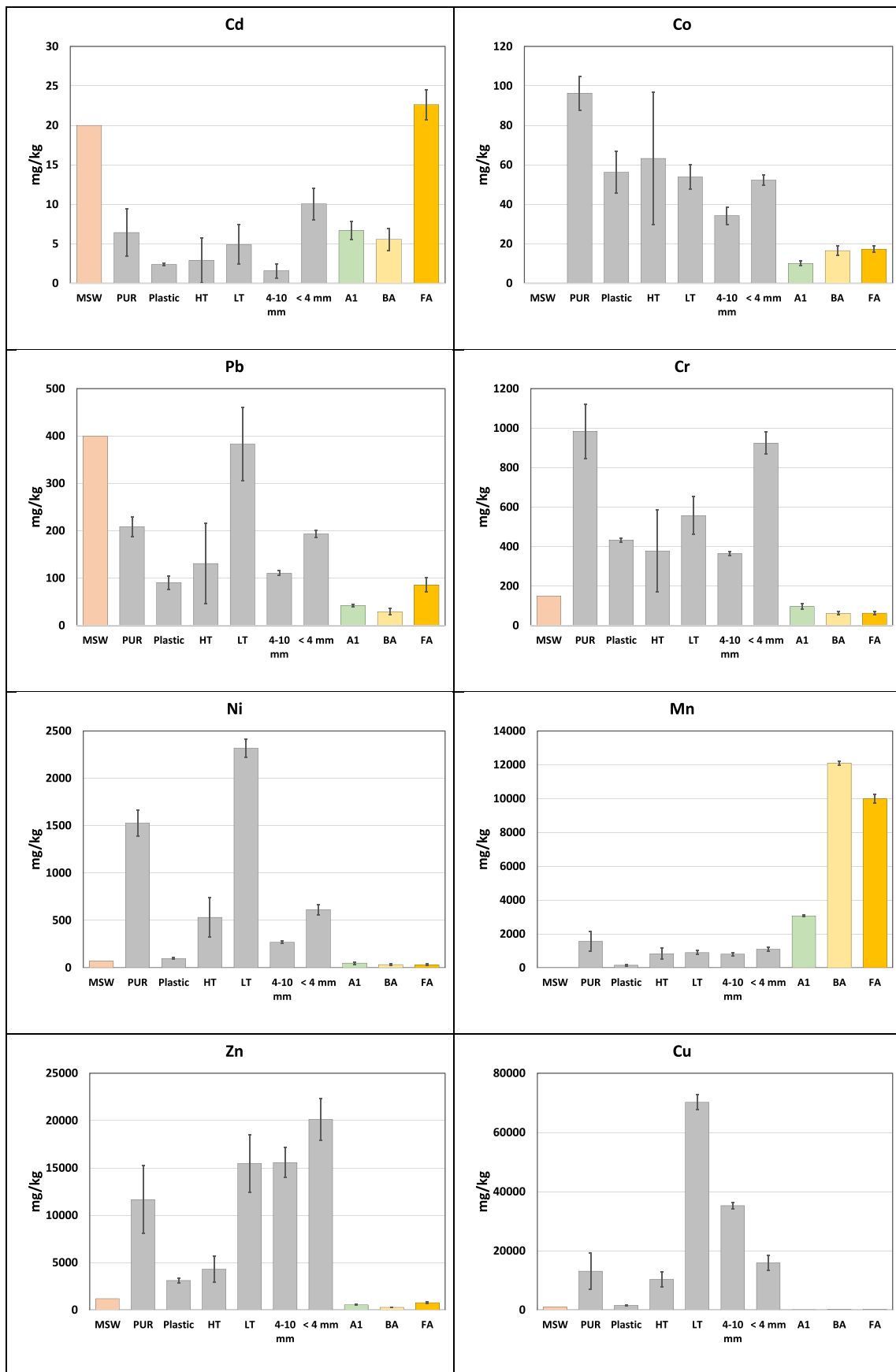


Fig. 6. Results of the ashes characterization (Cd, Co, Cr, Cu, Mn, Ni, Pb, Zn). The ashes came from the thermal valorization plant (MSW), the ASR foam rubber (PUR), the ASR heavy textile (HT), the ASR light textile (LT), the pellet type n.1 (A1), the pellet type n.2 bottom (BA), the pellet type n.2 fly (FA).

source for primary mining, through hydrometallurgical processes (Granata et al., 2011; Ferella et al., 2015). Remaining ashes could be landfilled or used as construction material (Lee, 2007).

Zinc was not included in EN 15359 standard for SRF assimilation, however its content in the ashes coming from the ASRs was in the order of 1–2% b. w., with the highest amounts found in the LT and fine fractions (<10 mm). The high amount of Ni in the foam rubber coming from the ASR samples collected in the 2017 sampling campaigns (see Table 2), determined high concentrations, in the order of 1500 mg/kg, also in the foam rubber (PUR) ashes. The content of Cr ranged from approx. 400 to 1000 mg/kg, depending on the ASR fraction, and was from 2 to 5 times higher than the amount found in MSW or wood ashes. Conversely, the content of Pb was found of the same order of magnitude, that is approx. 400 mg/kg, or, for some ASR fractions (i.e. heavy textile, plastic and foam rubber) even lower, of that of the ashes coming from the MSW thermal valorization plant. Higher Pb concentrations, in the order of 1000–1500 mg/kg, were reported by Huber et al. (2019) for the finest fractions (<8 mm) of MSW incineration bottom ash. Even Cd seemed not to be a concern. The content of Cd in the ashes generated from the ASR fractions was, on average, below 10 mg/kg, that is of the same order of magnitude of the Cd found in the pellet wood type 1 ashes (A1) and in wood pellet type 2 bottom ashes (BA). The Cd content found in the MSW ashes and in the pellet wood type 2 fly ashes (FA) was from 2 to 4 times higher. Cd concentration in the fly ashes, with respect to the bottom ashes, generated from the combustion of ASRs at a full-scale plant was well documented by Mancini et al. (2014). Finally, the Mn content of the ASR ashes was from 1.5 to even 10 times lower than that observed in wood pellet type 1 and wood pellet type 2, respectively. In general, the metals' content of the ashes generated from the combustion of the light ASR fractions was in line with the values reported in previous studies (Mancini et al., 2014; Lee et al., 2015), even if those studies considered the ASR in its entirety and it was combusted at full scale treatment plants.

4. Conclusions

Even if Europe achieved reuse-recycling rates in the order of 90%, most European countries still fail to comply with the targets of 2000/53/EC Directive. The assimilation of ASRs with an SRF could contribute to increase the recycling rate of ELVs. However, the results obtained in this study, after the characterization of ASR samples collected in four sampling campaigns, demonstrated that.

- The sole fraction of plastic extracted from light ASR had a quality which was compliant with EN 15359 standard and DM 22/2013 and could consequently be assimilated to an SRF-fuel; the abundance of plastic into ASRs, in the order of 15–20% b. w., could increase the reuse-recycling rate by approx. 2.4–3.3%, thus rising it from 82.2% to 84.6–85.4%;
- Heavy textile and foam rubber had an adequate NCV and compliance with Cl and Hg parameters, however the content of Cu, Ni and Sb (this last for heavy textile) made them not suitable to be assimilated to an SRF-fuel. However, the thermal valorization of those fraction could contribute to increase the reuse-recovery rate, thus targeting the EC goal (95%) and reducing the amount of light ASR which is still disposed in landfill;
- The removal of pieces of foam rubber of less than 40 mm, through a sieving operation, could improve the quality of the material, but that treatment was not sufficient to lower the content of Cu and Ni below the threshold values fixed by DM 22/2013;
- A model describing the metal content of foam rubber particles as a function of their particle size was developed and validated. It predicted that particle sizes of more than 200 mm were necessary to lower the Cu and Ni content of foam rubber below the threshold values fixed by DM 22/2013;

- The content of Cu and Zn in the combustion ashes made them of interest for metal recovery through hydrometallurgical processes. The content of some of the most dangerous heavy metals, namely Cd and Pb, was of the same order of magnitude, or even less, of that found in the ashes coming from the thermal valorization of MSW. The Mn content of the ASR ashes was from 1.5 to even 10 times lower than that observed in commercial wood pellets.

CRedit authorship contribution statement

Barbara Ruffino: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Mariachiara Zanetti:** Conceptualization, Funding acquisition, Project administration, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envres.2024.118131>.

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