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Pretreatment of automobile shredder residue (ASR) for fuel utilization

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Abstract

Automobile shredder residue (ASR) was pretreated to improve its quality for fuel utilization. Composition analysis revealed that ASR components could be classified into four groups: (1) urethane and textile—light fraction and combustibles containing low levels of ash and Cl; (2) plastics and rubber—light or heavy fraction and combustibles containing high levels of Cl; (3) metals and electrical wire—heavy fraction and incombustibles, and (4) particles smaller under 5.6 mm with high ash contents.

Based on these results, we successively performed sieving to remove particles smaller than 5.6 mm, float and sink separations to reject the heavy fraction and plastics and rubber containing Cl, thermal treatment under an inert atmosphere to remove Cl derived from PVC, and char washing to remove soluble chlorides.

This series of pretreatments enabled removal of 78% of the ash and 91% of the Cl from ASR. Sieving using a 5.6-mm mesh removed a considerable amount of ash. Product quality was markedly improved after the float and sink method. Specifically, the sink process using a 1.1 g cm^{-3} medium fluid rejected almost all rubber containing Cl and a large amount of PVC. The remaining Cl in char after heating at $300 \text{ }^{\circ}\text{C}$ under an inert atmosphere and washing was considered to be present as insoluble chlorides that volatilized at temperatures above $300 \text{ }^{\circ}\text{C}$. Based on a tradeoff relationship between product quality and treatment cost, ASR may be utilized as a form of refuse plastic fuel or char.

Key words: Ash and Cl removal, automobile shredder residue (ASR), fuel utilization, thermal recycling.

1. Introduction

With the development of automobile industry and the increase of automobile production, treatment and disposal of end-of-life vehicles (ELVs) have recently become important issues. More than 30 million ELVs are generated each year in the USA, the EU, and Japan (Staudinger and Keoleian, 2001; Péra et al., 2004; Kurose et al., 2006). At present, ELVs are dismantled and valuable parts are preferentially recovered. The chassis is then shredded to reduce volume and to recover ferrous and non-ferrous metals. The remaining fluff-like material, referred to as automobile shredder residue (ASR), comprises 20–30 wt% of an ELV and is sent to landfills (Rausa and Pollesel, 1997; Zolezzi et al., 2004; Matsuto et al., 2007). Due to the difficulties in securing new landfill sites and rising disposal fees, many countries have instituted new recycling policies to reduce the amount of ASR sent to landfill. The EU has established European Directive 2000/53/CE, which states that by January 1, 2015, the ELV reuse and recovery rate must be at least 95% by average weight per vehicle and year (European Commission, 2000). The Japanese government also instituted a law requiring more than 95% of each ELV to be recycled by the year 2015. To meet this requirement, more than 70% of ASR must be recycled (Japanese Ministry of the Environment, 2003).

ASR is difficult to recycle due to its heterogeneous composition, but thermal or feedstock recycling may be feasible due to its high plastic contents. Previous studies have examined incineration, input to cement kiln, pyrolysis, and gasification of ASR for such purposes (Braslaw et al., 1991; Day et al., 1996; Rausa and Pollesel, 1997; Horii and Iida, 2001; Roy and Chaala, 2001; Pasel and Wanzl, 2003; Zolezzi et al., 2004). The Cl and ash contents of ASR may be critical factors in these processes; 1 g of ASR has been reported to contain 10–40 mg Cl and 0.2–0.5 g ash (Day et al., 1996; Roy and Chaala, 2001; Tai et al., 2006). Material breakdown of American automobiles manufactured in 1980, 1990, and 1995 revealed that PVC usage was 7.6–9.4 wt% of all plastics and non-tire rubber (Braslaw et al., 1991; Staudinger and Keoleian, 2001). Flame retardant in plastics, varnishes, and paints could also be sources of Cl (Vassilve and Braekman-Danheux, 1999; Roy and Chaala, 2001).

Cl contents greater than 3 mg g⁻¹ may cause serious corrosion and fouling during combustion (Chen and Pagano, 1986). In addition, high ash content lowers heating values and increases the cost of final residue disposal; it can also cause corrosion, fouling, deposition, slagging, sintering, and agglomeration in thermal treatment system (Arvelakis and Koukios, 2002). Ideally, the Cl and ash contents of ASR would be reduced using a simple pretreatment prior to thermal or feedstock recycling.

The objectives of this study were to characterize ASR and develop pretreatment methods to improve its quality for fuel utilization. We characterized ASR by investigating its physical and chemical composition, and identified a strategy for pretreatment. We evaluated the product yield, combustibles, ash, Cl, and high heating value (HHV) in recovered product after each pretreatment.

2. Materials and Methods

2.1. Automobile shredder residue

We obtained about 29 kg of ASR in July 2006 from an ELV disassembly and shredding plant, where ELVs were manually disassembled and engines, gasoline, batteries, and air bags were recovered. The remaining chassis were then crushed, shredded, and sent through air classification, an aluminum separator, and manual sorting to recover recyclable metals. The ASR was quartered three times in the laboratory to obtain a representative sample, which was then dried at 60 °C for further experiments.

2.2. Composition analysis

The dried ASR sample was screened using a 5.6-mm sieve to separate the fraction that could not be sorted manually. ASR particles larger than 5.6 mm were sorted into seven components: urethane, textile, rigid plastic, soft plastic, rubber, electrical wire, and metal. Each component was then shredded to a size less than 2 mm using a cutting mill to measure ash, volatile matter (VM), fixed carbon (FC), and Cl contents.

Based on proximate analyses of coal and coke (JISC, 2004), we determined ash content by measuring residual weight after heating 1 g of the sample at 800 °C for 2 h. We determined VM content using weight loss during heating of a 1-g sample in a

lidded crucible at 900 °C for 7 min. FC was calculated using the follow equation: ash + VM + FC = 1. Cl content was measured using an incineration method with a tubular quartz reactor and absorption bottles (JISC, 1999). We placed approximately 1 g of the sample on a boat and loaded it into an 800 °C tubular reactor for 30 min; pure oxygen was used for complete combustion at a flow rate of 0.2 L min⁻¹. The generated gas was scrubbed as it passed through distilled water in absorption bottles. Volatile Cl (Cl_V) was determined using the sum of chloride ions dissolved in distilled water and chlorides adhering to the tubular reactor wall. To extract chlorides fixed in the residue, 50 mL of a 20% nitric acid solution were added to the combustion residue and then heated at 100 °C for 1 h. After the mixture was filtered, chloride ions in the filtrate were determined as residual Cl (Cl_R). Total Cl (Cl_T) was determined by the sum of Cl_V and Cl_R (Hwang et al., 2006). Chloride ion concentration was measured by the mercuric thiocyanate method (JISC, 2002), using an absorption spectrophotometer (U-1101; Hitachi Co.) at 460 nm.

3. Results and Discussion

3.1. Physicochemical composition of ASR

Table 1 lists the physical composition of ASR and the chemical properties of individual ASR components. Based on these data, Fig. 1 presents the distribution of ash, FC, VM, Cl_V, and Cl_R in 1 g of ASR. The ASR was composed mainly of plastics, rubber, and urethane, but it still contained unrecovered metals and electrical wire

during air classification and magnetic separation. Particles smaller than 5.6 mm comprised 0.18 g g^{-1} of the ASR, and included soil, grit, glass, and miscellaneous materials. As shown in Fig. 1a, metals and particles smaller than 5.6 mm had a considerable influence on the ash content of ASR. Urethane, textiles, plastics, and rubber increased the combustibles ratio (VM + FC). However, plastics and rubber should be recovered selectively because these materials have high Cl contents (Fig. 1b); soft plastics had the highest Cl content (12.3 mg g^{-1} ASR), followed by rubber (6.9 mg g^{-1} ASR) and rigid plastics (6.0 mg g^{-1} ASR). Electrical wire has a high Cl content, but due to its small weight ratio in ASR, the content was relatively low (0.12 mg g^{-1} ASR).

3.2. Strategy for ASR recovery and pretreatment methods

We used the physicochemical composition of ASR, shown in Fig. 1, to classify its quality using the following categories. *G1* refers to urethane and textiles—a light fraction and combustibles containing low ash and Cl. *G2* refers to plastics and rubber—a light or heavy fraction and combustibles containing high Cl. *G3* refers to metals and electrical wire—a heavy fraction and incombustibles. *G4* refers to particles smaller than 5.6 mm with high ash contents.

Improving the quality of recovered ASR requires recovering *G1* and *G2*, but Cl must first be removed from *G2*. *G3* and *G4* should also be removed from ASR. *G4* can be removed using a sieve, and because *G3* is heavier than *G1* and *G2*, it can be removed using density-based separation, such as the float and sink method. By investigating the weight ratios and compositions of float and sink fractions for *G1*, *G2*, and *G3* in fluids with various densities, it is possible to determine the appropriate fluid

density to recover *G1* and *G2* selectively and to remove *G3*. The recovered ASR can be washed or heated at low temperatures in an inert atmosphere to reduce its Cl content.

Table 2 lists the product yield and quality after a series of pretreatments. Product yield represents the weight ratio of recovered ASR to untreated ASR. Combustibles (FC + VM), ash, and Cl content were presented as quality items.

3.3. Removal of particles smaller than 5.6 mm via sieving

We could easily remove *G4* using a 5.6-mm sieve. Calculations based on the data shown in Table 1 revealed that ash content decreased from 0.37 to 0.31 g g⁻¹ whereas Cl content increased from 29 to 34 mg g⁻¹ with weight reduction (see Table 2).

3.4. Reduction of Cl and ash content via float and sink separation

During the next stage, we subjected ASR components larger than 5.6 mm to float and sink separation to remove high levels of ash and Cl-containing components, such as PVC in *G2* and incombustibles in *G3*. The fluid began with distilled water (density of 1.0 g cm⁻³). Based on the specific gravity of PVC, we added calcium nitrate tetrahydrate (Ca(NO₃)₂·4H₂O) to the distilled water to make the fluid density with 1.1 and 1.4 g cm⁻³. Individual ASR components larger than 5.6 mm were immersed in the fluid medium with a liquid to solid ratio (L/S) of 15, stirred, and allowed to settle for 10 min. Floating and sinking materials were collected separately

and washed with 3 L of tap water and 1 L of distilled water and then dried overnight at 80 °C.

At a fluid density of 1.0–1.4 g cm⁻³, almost all *G1* components floated, and most *G3* components sank. Therefore, the quality of ASR recovered using float and sink separation depends on the behavior of *G2* in a given density of fluid medium.

Figure 2 presents the distribution of rigid plastics, soft plastics, and rubber by density range (1.0–1.4 g cm⁻³) and presents breakdown by combustibles, ash, Cl_V, and Cl_R. The fraction < 1.0 g cm⁻³ is the float in a fluid with a density of 1.0 g cm⁻³ and the fraction > 1.4 g cm⁻³ is the sink in a fluid with a density of 1.4 g cm⁻³ medium fluid, respectively. We determined the weight of the fraction with a density between 1.0 and 1.1 g cm⁻³ by subtracting the weight of float at 1.0 g cm⁻³ from that of the float at 1.1 g cm⁻³. As shown in Fig. 2, most combustibles were distributed in the fraction with a density less than 1.1 g cm⁻³, whereas Cl was found mainly in the fraction with a density greater than 1.4 g cm⁻³.

Table 2 presents the yield and quality of recovered ASR using fluid media with densities of 1.0, 1.1, and 1.4 g cm⁻³. The combustible recovery rate was lowest using a fluid medium with a density of 1.0 g cm⁻³. There were no marked differences in production yield or quality using fluids with densities of 1.1 and 1.4 g cm⁻³. However, use of a fluid with a density of 1.1 g cm⁻³ may minimize costs related to chemicals and result in slightly higher quality ASR recovery in comparison to a fluid with a density of 1.4 g cm⁻³.

3.5. Dehydrochlorination via thermal treatment in an inert atmosphere

Even after float and sink separation, 1 g of the recovered ASR still contained 9.3 mg g⁻¹ Cl_T and 8.9 mg g⁻¹ Cl_V (Table 2). As almost all Cl from rubber was removed as the sink at 1.1 g cm⁻³, the remaining Cl in the float at a fluid density of 1.1 g cm⁻³ was probably derived from plastics (Fig. 2c). If recovered ASR still contains Cl compounds derived from PVC, they may be volatilized by heating under inert conditions. Dehydrochlorination reaction occurs in the temperature range of 200–360 °C during pyrolytic degradation of PVC (Miranda et al., 1999). We conducted a preliminary experiments using a small tubular reactor (D × L = 28 mm × 500 mm); when we heated a 1 g reference sample (PVC pipe:PVC film = 10:1) at 300 °C for 30 min under a stream of nitrogen gas at a flow rate of 100 mL min⁻¹, the entire Cl content was transferred to the gas phase.

Individual components that floated in the 1.1 g cm⁻³ fluid were shredded and mixed again based on weight ratio. We placed approximately 20 g of the sample on a boat and loaded it into a tubular ceramic reactor (D × L = 60 mm × 1000 mm), which was heated at 300 °C for 60 min under a stream of nitrogen gas at a flow rate of 1.4 L min⁻¹. After this thermal treatment, we collected and weighed the char (the remaining solid residue on the boat). The char yield was about 86% and Cl content decreased to 7.2 mg (Table 2). About 33% of the Cl remaining in the float in the 1.1 g cm⁻³ medium fluid was removed by thermal treatment at 300 °C in an inert atmosphere, and so this Cl was probably derived from PVC. In contrast, the Cl remaining in the char may have come from additives to textiles and plastics, such as vanishes, paints, and pigments.

3.6. Removal of soluble chloride via washing

If the Cl compounds in char are in the form of soluble chloride, this may be dissolved in water during the washing procedure. We pulverized char into particles smaller than 1 mm, which were then washed with distilled water in L/S = 10 with shaking at 150 rpm for 20 min. The mixture was filtered using a vacuum pump, and we then recovered dewatered char. This entire procedure was repeated four times, and reduced the Cl content of char from 7.2 to 5.4 mg g⁻¹, representing a 24% Cl removal rate (Table 2).

4. Evaluation of ASR pretreatment

Figure 3 illustrates our entire ASR pretreatment process and the mass balance of combustibles, ash, and Cl in untreated ASR was 1. We removed one third of all ash in the ASR by separating particles smaller than 5.6 mm. Float and sink separation using a 1.1 g cm⁻³ fluid removed 46% of ash and 78% of Cl from the ASR. Another 9% of Cl was removed during thermal treatment at 300 °C and washing. On average, 11% of combustibles were lost during every pretreatment from sieving to thermal treatment. At the end of our series of pretreatments, we recovered 67% of combustibles and removed 14 and 9% of ash and Cl from the ASR, respectively.

Figure 4 shows the various product yields and qualities obtained by pretreatment. We estimated ASR HHV using HHV data from individual components as listed in Table 1, and calculated char HHV using the following equation: $HHV_{Char} [MJ/kg] = (32.8 \times FC + 19.6 \times VM)$ (Hwang et al., 2007). By the end of the series of pretreatments, product yield had decreased to 0.5 g g⁻¹ ASR. Sieving using a

5.6-mm mesh is a simple method to remove ash. Product quality was markedly improved by removing the fraction $> 1.1 \text{ g cm}^{-3}$, and HHV increased to 29 MJ kg^{-1} . The quality standards for refuse paper and plastic fuel (RPF) as a substitute for coal are $\text{HHV} > 25 \text{ MJ kg}^{-1}$, $\text{ash} < 0.7 \text{ g g}^{-1}$, and $\text{Cl} < 3 \text{ mg g}^{-1}$ (Japan RPF Association, 2004). The float at 1.1 g cm^{-3} satisfied the HHV criterion, but the ash and Cl contents were still high.

We removed Cl from float at 1.1 g cm^{-3} using a dehydrochlorination reaction during thermal treatment at $300 \text{ }^\circ\text{C}$ in an inert atmosphere. Despite considerable loss of combustibles during heating (Fig. 3), the process removed a relatively small amount of Cl derived from PVC. This result indicates that a large amount of PVC had already been removed during float and sink separation, and that the Cl in float at a fluid density of 1.1 g cm^{-3} was mainly derived from additives, such as varnishes, paints, and pigments, rather than from PVC. The Cl that remained after char washing was regarded as insoluble chloride, which is not volatilized at temperatures below $300 \text{ }^\circ\text{C}$.

5. Conclusions

Feedstock and thermal recycling may be the most feasible method for raising the recycling rate of high caloric but heterogeneous materials, such as ASR. Under these conditions, Cl and ash contents become critical determinants of whether waste-derived fuel can be utilized in a plant.

ASR is comprised of about 70 wt% combustibles, such as urethane, textile, plastics, and rubber, with the other 30 wt% was composed of metals, electrical wire, and particles smaller than 5.6 mm. We used the physicochemical composition of ASR components to classify them into four groups and then established a strategy for pretreatment. Sieving using a 5.6-mm mesh removed one third of the ash from ASR; this was a simple and effective method for removing ash. Float and sink separation removed a considerable quantity of incombustibles and a large fraction of the ash and Cl; a sink using a fluid density of 1.1 g cm^{-3} removed 46% of ash and 78% of Cl from the ASR. Almost all Cl derived from rubber and a large quantity of PVC were removed during float and sink separation, whereas thermal treatment and char washing did not markedly affect Cl levels.

ASR can be utilized in the form of RPF or char depending on requirements. The benefits of using thermal treatment to reduce Cl are still debatable due to the financial and energy requirements. Nevertheless, in addition to removing Cl, the process yields a char with many advantages: improved quality control, ease of handling, and improved substitutability in existing coal-fired power boilers. Thus, product choice will be based on a tradeoff between product quality and treatment cost.

This study focused primarily on pretreatment of ASR from the perspective of fuel utilization and thermal recycling. To achieve the required 70% ASR recycling rate, further research is needed to address the recovery of metals from the rejected portion.

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Table 1. Physical composition of ASR and the chemical properties of individual ASR components

Component	Weight ratio ^a (g g ⁻¹ ASR)	Component properties ^b						
		VM (g g ⁻¹)	FC (g g ⁻¹)	Ash (g g ⁻¹)	HHV ^c (MJ kg ⁻¹)	Cl _V (mg g ⁻¹)	Cl _R (mg g ⁻¹)	Cl _T (mg g ⁻¹)
Urethane	0.11	0.76	0.03	0.20	21	9.4	0.4	9.8
Textile	0.09	0.65	0.04	0.31	23	5.9	0.4	6.3
Rigid plastics	0.32	0.85	0	0.15	36	17	2	19
Soft plastics	0.05	0.85	0.04	0.10	21	228	17	245
Rubber	0.13	0.60	0.16	0.24	26	42	11	53
Electrical wire	0.01	0.36	0.03	0.60	8.4	95	28	123
Metals	0.11	0	0	1.00	0	0	0	0
< 5.6 mm	0.18	0.06	0.30	0.64	8.1	5.2	1.2	6.4

^a: n = 3.

^b: per 1 g component.

^c: from Matsuto (2004).

Table 2. Product yield and quality of recovered ASR by pretreatment

Pretreatment	Product	Product yield ^b (g g ⁻¹ ASR)	Product quality ^c				
			Combustibles (g g ⁻¹)	Ash (g g ⁻¹)	Cl _V (mg g ⁻¹)	Cl _R (mg g ⁻¹)	Cl _T (mg g ⁻¹)
No pretreatment	ASR	1.00	0.63	0.37	26	3.5	29
Sieving using a 5.6-mm mesh	ASR > 5.6 mm	0.82	0.70	0.31	30	4.1	34
Float and sink separation	Float at 1.1 g cm ⁻³	0.58	0.86	0.14	8.9	0.4	9.3
	Float at 1.0 g cm ⁻³	0.43	0.84	0.16	9.1	0.6	9.7
	Float at 1.4 g cm ⁻³	0.60	0.85	0.15	9.3	0.3	9.6
Thermal treatment at 300 °C under an inert atmosphere	Char ^a	0.50	0.84 ^d	0.16	6.5	0.8	7.2
Washing (four times)	Washed char	0.50	0.84 ^d	0.16	5.1	0.4	5.5

^a: Produced via floating in a 1.1 g cm⁻³ medium fluid.

^b: g per 1 g untreated ASR.

^c: mg or g per 1 g product.

^d: = 0.82 (VM) + 0.02 (FC).

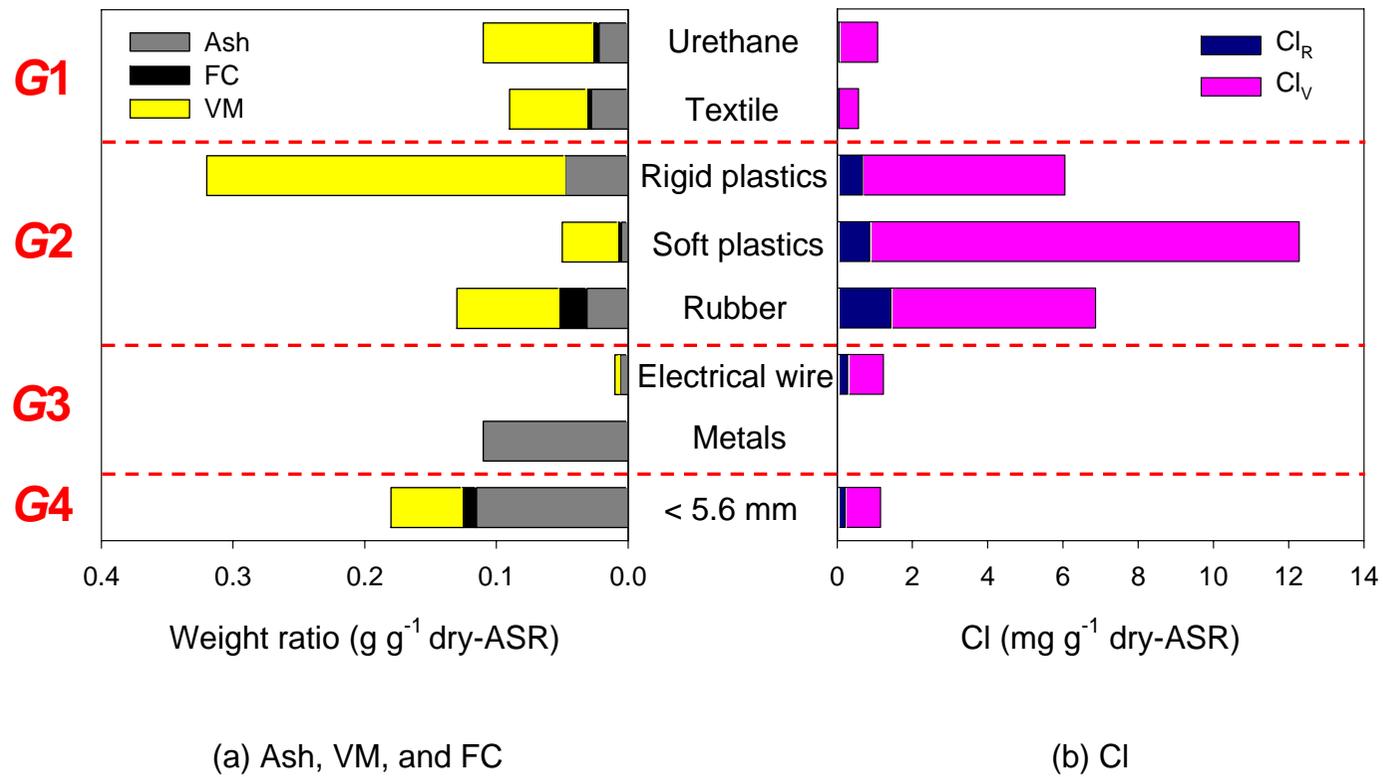


Fig. 1. Physicochemical composition of 1 g ASR.

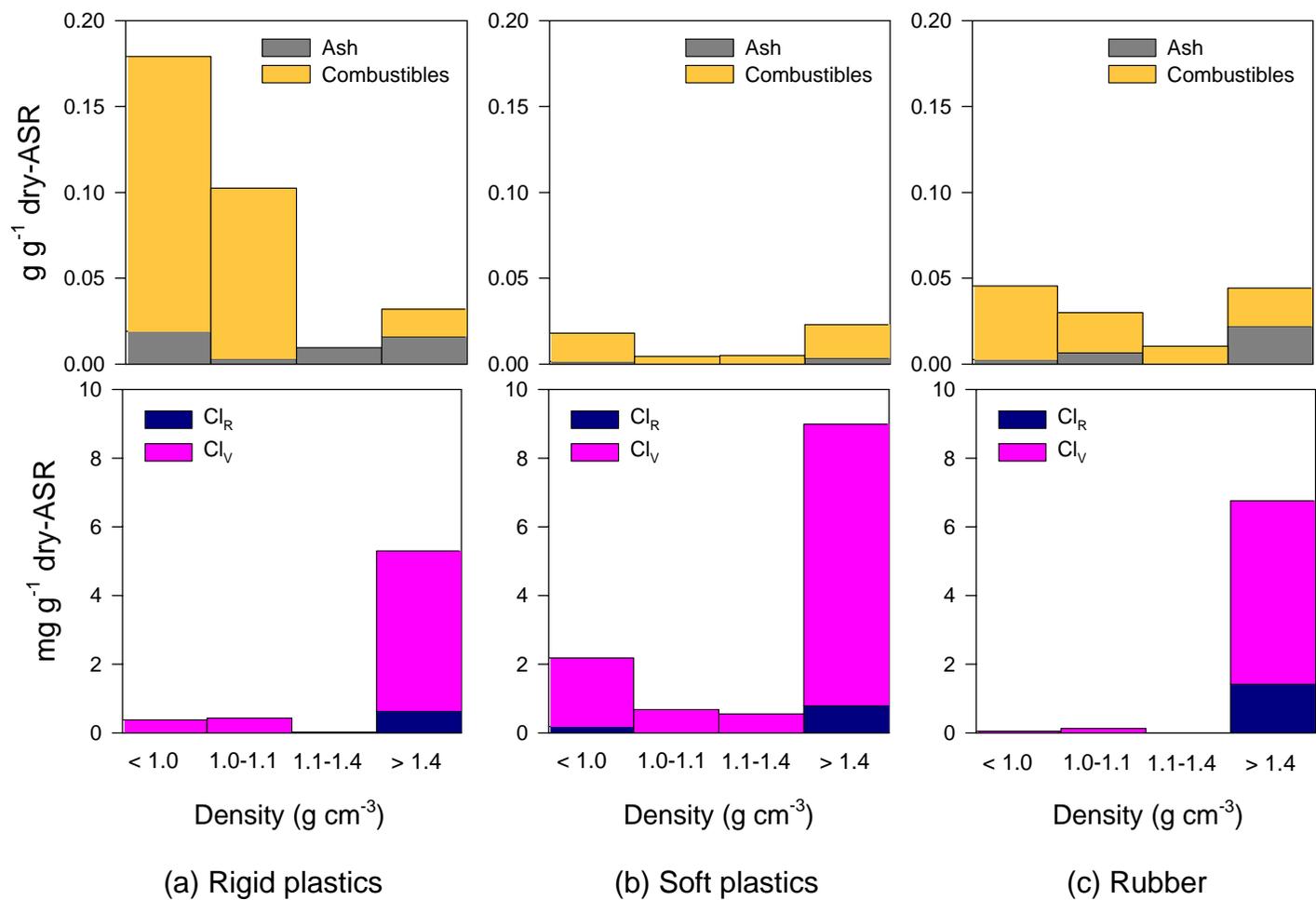


Fig. 2. Distribution of combustibles, ash, Cl_V , and Cl_R in rigid plastics, soft plastics, and rubber classified by density range.

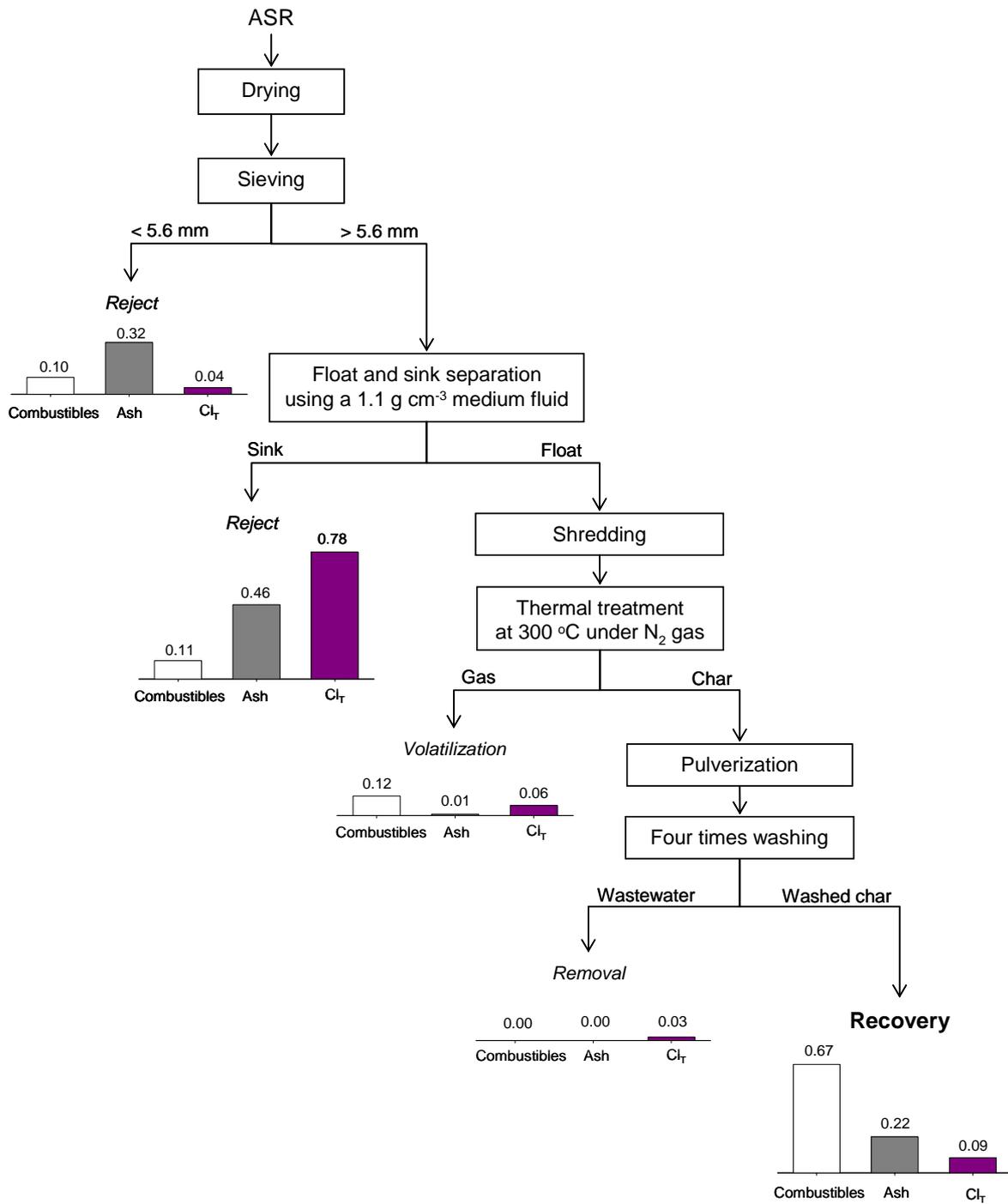


Fig. 3. Entire ASR pretreatment process and the mass balance of combustibles, ash, and Cl of untreated ASR is 1.

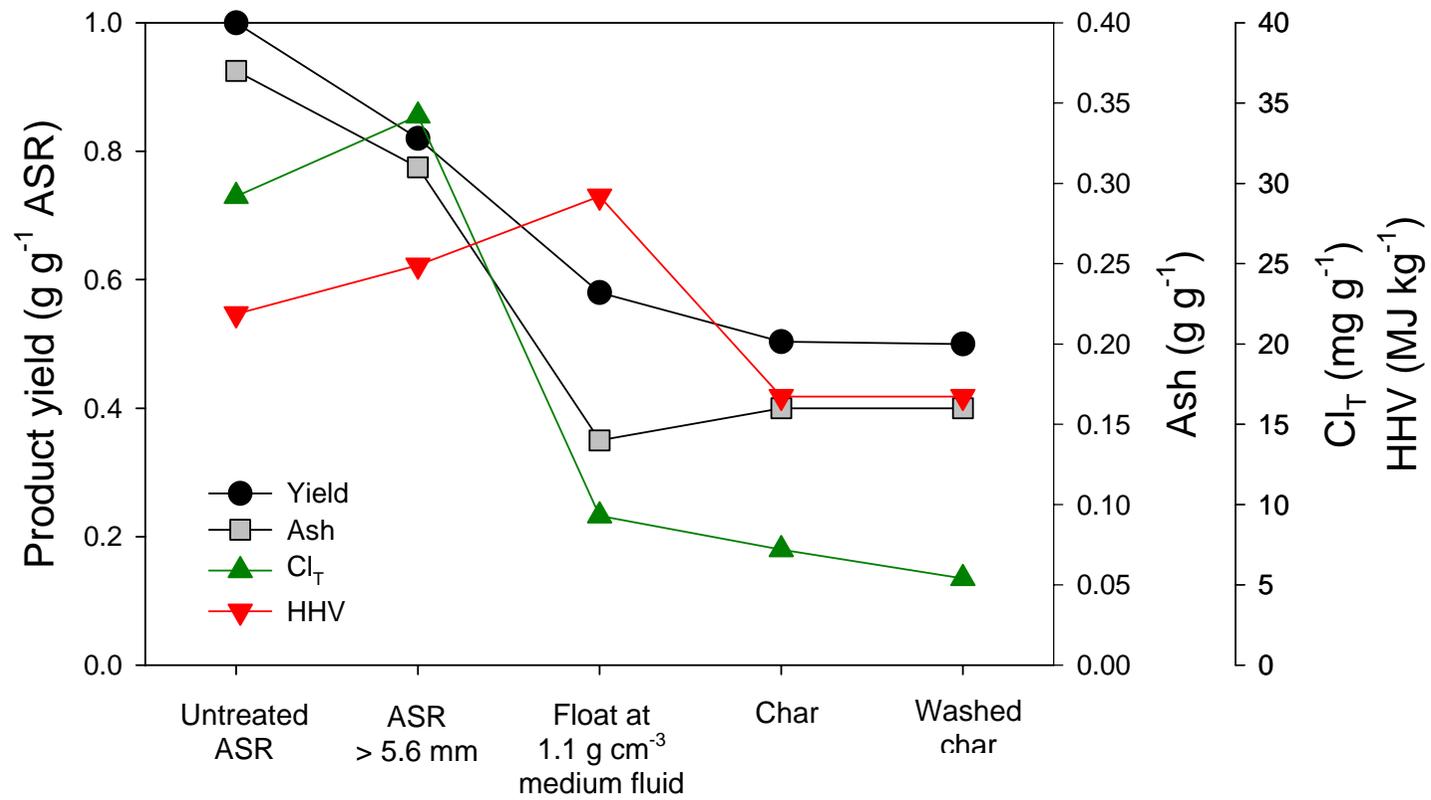


Fig. 4. Variation in product yield and quality by pretreatment.