Recycling of aluminium from mixed household waste

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Abstract

Aluminium is used in packaging due to its density, strength and preservative capabilities. This paper outlines a methodology to evaluate the recyclability of aluminium waste, where parameters of scrap type, pre-treatment and remelting conditions are alternated in a factorial design, with measurements of yield and analysis of metal quality. In preliminary experiments three aluminium waste materials were evaluated for recyclability. The methodology was tested on one of the waste materials from mixed household waste, named Cans, and will be described in detail. Cans was decoated at 300 °C and 550 °C. The decoated material was then remelted, 4 separate additions of ~ 2 kg per experiment, by submerging the sample material in molten aluminium. Porosity, oxide inclusion content and alloy composition were analysed to determine metal quality. The experimental results show that the yield of remelting, dross formation relative to melted sample material and the alloy composition of the remelted metal is dependent on decoating temperature. In conclusion: Cans is recyclable, with good quality and yield, when decoated before melting. Decoating at 550 °C improves metal yield and oxide inclusion content compared to decoating at 300 °C, however it also reduces magnesium content in the resulting alloy. Hopefully the methodology was useful for assessing the recyclability of an aluminiumrich waste and for evaluating the effects of various process parameters during recycling on metal yield and quality.

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Introduction

Aluminium is used in packaging due to its density, strength and preservative capabilities. Even with the best efforts, with major collection and sorting systems in place, will there inevitably be aluminium that ends up in a fraction called municipal solid waste, now often denoted mixed household waste. As waste processors begin to sort their waste streams, large amount of aluminium waste will become available for recycling. This would be a natural response in Europe to meet the recycling requirements introduced in the directive (EU) 2018/851 of the European Parliament, which is an amendment to the directive 2008/98/EC on waste (in which 50 % of materials such as metal, glass, cardboard and plastics passing through households need to be re-used or recycled, increasing by 5% every 5 years until 65% in 2035).

Picking analyses made with several hundreds of kilograms of mixed household waste from different Norwegian regions estimated a metal waste content (described as mostly aluminium waste) of approximately 1% - 2%. [2], [3] Internal picking analyses at IVARs sorting plant in Stavanger gave aluminium contents of 1.3 %. Studies from *Politecnico di* Milano reported Aluminium waste contents of 0.73 % and 1.63 %. [4]–[6] A minimum aluminium waste content of 1 % is therefore a reasonable and conservative estimate for Norwegian mixed household waste.

A methodology was developed to assess the recyclability of such a waste, sourced from a sorting plant in Norway, as a natural first step in developing its recycling. This paper details the methodology, the sample material and some of the results from these experiments.

Theory

Hydrocarbons, such as those found in plastics, coatings, lacquer and food stuffs upon heating break down in endothermic scission reactions producing smaller molecules such as CH_4 , CO, CO_2 , H_2O , tar and oxygenated oils depending on the balance between carbon, hydrogen and oxygen in the material. If this heating is done in atmosphere with oxygen CO_2 and H_2O will be produced from a thermodynamic perspective, but this depends on a surplus of oxygen. In an incomplete combustion scenario with an oxygen-depleted atmosphere one can expect to find a mixture of all these molecules. [7] Increasing in the order CH_4 , CO, C_4H_{10} , CO_2 have been shown to significantly increase oxidation of an aluminium melt compared to 5% O_2 in Argon or pure Argon atmospheres [8]. Incomplete decoating where remnant pyrolysis products are left on aluminium samples have also been shown to reduce metal coalescence, increase dross formation and reduce yield compared to complete decoating process in which the coating and plastics are completely removed. [9] The elemental properties listed in table 1 will be used in discussions of alloying contents in the recycled metal.

Table 1: Melting point, vapour pressure and formation enthalpy of oxide per anionic oxygen for select elements. [10], [11] at the highest decoating temperature.

Element	Melting Point	Vapour Pressure at	ΔH_f° of oxide per O	
	[°C]	527 °C [Pa]	[kJ·mol ⁻¹]	
Al	660	3.05 · 10 ⁻¹⁰	-558.6	
Mg	650	2.15 · 0¹	-601.6	
Pb	327	6.18 · 10 ⁻³	-219.0	
Sn	232	1.26 · 10 ⁻⁹	-280.7	

Materials

The mass fraction of aluminium in a packaging depends on, and can be categorized after, the function aluminium plays in the item. This categorization is useful as it coincides with how easy the item can be recycled, including sorting, pre-treatment and remelting, decreasing with increasing order: [1]

- Category 1 Structural: Up to 98 % aluminium by mass. Items like cans for beverages and canned foods, trays for roasting fish and chicken, aluminium tubes for cosmetics and food spreads and coffee pods. Typically contains significant amounts organic contamination.
- Category 2 Functional: Smaller pieces of aluminium attached to larger pieces of other packaging materials, often designed to tear or be removed upon opening, used to seal toothpaste tubes, pill cards and many food items. Typically attached to other materials such as plastics, papers.
- Category 3 Laminar: Aluminium rolled into thin foil used in laminate with organics such as plastic, lacquer (which could also contain metal oxides), paper and glue. The aluminium in such foil is typically thinner than 200 microns, and very difficult to separate out.

Packaging in a higher category will score lower in the following properties:

- 1. The amount of aluminium in the packaging;
- 2. the fraction of the aluminium in mixed household waste that stems from this category of packaging;
- 3. how easily the packaging is recycled, including sorting, pre-treatment and remelting; and
- 4. the yield and metal quality of remelted metal.

The sample material was retrieved from a mixed household waste sorting plant run in Stavanger by IVAR IKS which started operation in 2019. In the plant materials like paper, corrugated cardboard, metal and especially plastics are sorted from the mixed household waste through systems such as near infrared spectroscopy, mechanical sorting systems and magnetic extractors (including eddy current separators).

Unprocessed aluminium waste contains a large fraction of organics materials in the forms of foodstuffs and inherent co-materials such as plastics, coatings and lacquers. These cannot be part of a recycled metal and are typically removed thermally, whether this occurs during remelting or in a separate decoating step. The aluminium contains significant contents of alloying elements such as magnesium, iron and silicon. These are both a challenge and an opportunity: If utilized correctly recycled materials could require lower additions of alloying elements. Sometimes these alloying elements, which are difficult to remove from the aluminium melt, must be diluted with primary aluminium.

The Cans and Foils Sample Material

The aluminium waste fraction sorted out in Stavanger contained a mixture of categories 1, 2 and 3 aluminium waste. To create the *Cans* sample materials closed containers (which are operational hazards) and category 3 packaging was sorted out by hand at the plant before being sent to our facilities for experiments, as preliminary experiments had shown that category 3 packaging was not recyclable with the equipment and methods utilized in our remelting experiments. During hand sorting care was taken to not separate out pieces of packaging that looked difficult to recycle. This mixture of category 1 and 2 packaging was aptly named *Cans* after its main constituents, beverage cans and canned foods cans. Cans decoated at 300 °C (scisstion decoating) and 550 °C (combustion decaoting) is referred to as Cans 300 and Cans 550.

Cans consisted of a mixture of:

- Aluminium beverage cans;
- aluminium cans for canned foods;
- aluminium trays for fish fillets;
- partially empty tea lights containers;
- aluminium screw corks for bottles;
- used Aluminium coffee pods;
- aluminium tubes for spreadable cheese; and
- other Aluminium packaging items like trays for cream cheese or pet food.

The category 3 packaging shown in Figure 1 is sorted out would form a sample material dubbed *Foils*, which was used in preliminary experiments with *Cans* and UBC (Used beverage Cans).



Figure 1: The Foils sample material, composed of thin laminates with aluminium, lacquer and plastics.

Methods

The methodology is composed of the following key activities: [1]

- 1. Retrieval of aluminium sample material(s)
- 2. pre-treatment of the sample material(s)
- 3. Remelting of the sample material(s)
- 4. Analysis of the recycled sample material(s)

The methodology was utilized on the *Cans* sample material described above, where decoating temperature was the varied parameter. The experiments were performed on the biggest scale practically possible in a laboratory, with processing and equipment very similar to those encountered in industrial scenario, to produce as industrially relevant results as possible.

Pre-Treatment

Pre-treatment consisted of thermal decoating, compaction and sizing. Cans was decoated in batches of 6 troughs holding 1.5 kg to 4 kg of sample material each in a 50 kW resistance furnace. In trays shown in Figure 1 The batches were heated up to 300 $^{\circ}$ C or 550 $^{\circ}$ C and held at the temperature until decoating was completed (when the process stopped producing smoke). Decoating took about 90 minutes at 550 $^{\circ}$ C and 180 minutes at 300 $^{\circ}$ C.

After decoating the samples were turned in a 130 L cement mixer to remove ash and carbonaceous residue. The samples were turned at low speeds for 5 minutes each. The samples were then compacted at 35 MPa in an industrial metal press and cut into suitable pieces with a band saw. Prior to remelting experiments the samples, were heated to 150 °C to remove moisture.

Remelting

Remelting was performed in a copper coil-furnace with the crucible placed on top of a digital scale, allowing a continuous measurement of the crucible. In each experiment 4 additions of ~2 kg of material was melted by submersion into a pure molten aluminium heel. The melt was kept close to 800 °C, but temperatures varied greatly during melting due to the addition of cold sample material. After sample material was added, dross was skimmed with a perforated steel ladle, thereafter samples for reduced pressure testing were extracted and set to solidification in an evacuating chamber. Before addition, after addition, and after skimming, the mass of the crucible was measured. The mass measurements were always made when the furnace was unpowered, to avoid interference on the scales from the coil.

After skimming and sample extraction for reduced pressure testing in the 4th addition was completed, the whole melt was decanted into cast iron moulds for solidification, the ingot from the first mould filled was used to perform cold PoDFA analysis and optical emission spectroscopy analysis.

Yield and relative dross formations

The mass of sample additions to the crucible and dross skimmed was measured individually. These measurements were so similar to the masses calculated by comparing the measured masses of the crucible before and after addition, and after skimming, to such an extent that these calculated values were used as data for the *initial mass* - m_i of the sample added to the crucible, the *skimmed mass* - m_s of dross removed during skimming, and the *final mass* - m_f .

$$\mbox{Yield [\%]} = \mbox{$^{m_f}\!/_{m_i}$} * 100 \, \%$$
 Relative Dross Formation [\%] = $\mbox{$^{m_s}\!/_{m_i}$} * 100 \, \%$

As a result of the way final mass, initial mass and skimmed mass is measured, the following relation always holds true:

$$m_f + m_s = m_i$$

Data on temperature and duration was also gathered. All these data were measured for each addition in each experiment.

Reduced Pressure Test

Reduced pressure testing – RPT is performed to analyse the porosity of a liquid aluminium sample. The molten sample is poured into a sand mould solidified in a vacuum chamber kept at reduced pressure. The porosity of the sample, which is much larger than a sample solidified at atmospheric pressure, is assessed visually by splitting and polishing of the sample. The porosity of the sample will depend on the gauge pressure during solidification, the inclusion content of the sample, and the contents of dissolved elements like hydrogen which form pores during solidification.

Porous Disk Filtration Analysis (PoDFA)

PoDFA is a frequently used technique for determining the quantity of inclusions in an aluminium melt. Approximately 2 kg of the melt is poured into a preheated PoDFA crucible with a filter at the bottom. The crucible is placed over an evacuated chamber such that the melt is drawn through the filter. After cooling the filter is cut and polished for optical examination with SEM and micro probe. This allows characterisation of different inclusions types and the area of those types per kg of metal. In this project cold-PoDFA was utilised, Cold PoDFA means that the analysis was done with solidified samples at a later stage in the project,. In the *Aluminium melt cleanliness performance evaluation using PoDFA* an inclusion content in the melt below $0.05 \ mm^2/kg$ is considered very light, the best classification. [12]

Statistical Analysis

Several hypotheses were created and tested to prove a statistically significant differences between two sets of samples taken out of the populations of Cans 300 and Cans 550, assumed to be normally distributed. The quantifiable measurements such as yield and content of magnesium were then evaluated, mostly through visualisation using the Seaborn, a data visualisation library based on the Matplotlib library for Python.

The samples retrieved were assumed to be Student-T distributed, and the hypotheses claiming the measurements from both populations were equal, was tested with Minitab 2018 Statistical Software.

Results

Foils, Cans and UBC: Preliminary experiments

In preliminary experiments decoating at 550 °C and remelting was performed on Cans, Foils and UBC. These experiments showed that Foils, compared to UBC and Cans, was non-recyclable with the equipment and methods used. Foils lost 84 % of its mass during decoating, with a lot of ash trapped in the decoated metal. Foils had a negative yield: During remelting the sample oxidised more of the aluminium heel than the mass of the sample itself. Whilst Cans lost significant mass during decoating, the yield of UBC and Cans of 73 % and 74 % was comparable. Based on these results Cans was chosen as sample material. More details are available in the thesis [1]. During remelting of *Foils* the melt reached over 1300 °C suspected to be due to exothermic reactions.

Table 2: Results from preliminary experiments.

* Assumed increased decanted mass of 35 g based on 37 g bottom metal extracted after decanting remelted UBC.

	Decoating			Remelting					
	In	Out	Yield	Ash	Heel	Addition	Decanted	Yield	Dross
Foils	1114 g	174g	15.6 %	a lot	551g	176g	522g	-16.5%	a lot
UBC	1221 g	1175g	96.2 %	some	649g	218g	807g	72.5%	some
Cans	688 g	373g	54.2 %	little	564g	213g	722g*	74.2%	some

Pre-treatment

Figure 3 depicts Cans before and after decoating. The surface of Cans 300 was typically glossy black due to residual non-combusted pyrolysis products coating the sample surfaces. It had an odour similar to burnt food. there was no evidence of metal oxide formation on Cans 300 In contrast to Cans 550. The bottom of the troughs were filled with tar-like pyrolysis products to the extent of encapsulating flat aluminium pieces.

The surface of Cans 550 was matt grey and/or white, evidence of partial oxidation of the metal. It was completely free of odour. Melting of the sample material was evident from metal droplets, disintegrated aluminium packaging pieces and the samples in some troughs looking shrunken. The aluminium trays for chicken and fish especially were often partially melted.



Figure 3: Cans before and after decoating at 300 °C and 550 °C.

Cans 300 and Cans 550 exhibited a significant mass loss 43% during heat treatment, due to evaporation of water and combustion of organic content. These mass losses at 300 $^{\circ}$ C and 550 $^{\circ}$ C are were both similar to the mass losses in the preliminary experiments.

Table 3: Results from decoating, showing the summed masses of n batches of that temperature.

Temperature	n	Mass before	Mass After	Mass loss	Mass loss
300 °C	24	60.2 kg	35.3 kg	24.9 kg	41.4 %
550 °C	13	65.8 kg	37.3 kg	28.5 kg	43.3 %
Both	37	126.0 kg	72.6 kg	53.4 kg	42.4 %

The ash was not quantified but accounted for less than 5 % of sample mass. The ash from Cans 300 consisted mostly carbonaceous tar and char, whilst the ash from Cans 550 was darker grey, likely a mixture of the carbonaceous tar and char, and metal oxide.

Remelting.

10 remelting experiments were performed, 4 with Cans 300 and Cans 550, and 2 with pure aluminium ingot. Each experiment elapsed between 1 and 2 hours, depending on how quickly the samples melted. During remelting of Cans 300 flames often rose from the crucible, indicating production and combustion of flammable gas. During remelting of Cans 550 flames were sometimes observed, but at a drastically smaller scale.

Figure 4 shows how much metal was added, how much dross was skimmed out, and how much metal was kept after skimming of dross as yield, for each addition, for experiments 6 and 8. The Dross skimmed and Yield always sum to Metal added. No correlation was seen between yield and relative addition, which is the size relationship between how much mass is added and how much mass is already in the crucible. The *relative addition* varied between almost 100 % to 33 %.

	Cans 300	Cans 550
Average Yield [%]	53.8	65.3
Average Dross [%]	46.2	34.7
Total [%]	100.0	100.0

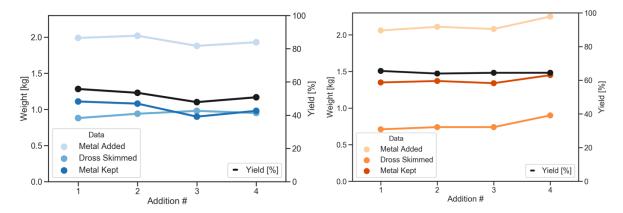


Figure 4: Calculated data for metal added, dross skimmed and yield [kg] for Cans 550 - experiment 6 and Cans 300 - experiment 8.

A yield of 53.8 % and 65.3 % was measured on average per addition for Cans 300 and Cans 550 respectively. The hypothesis that the yields are equal was tested using Minitab and rejected with a P-value of $3.6 \cdot 10^{-7}$, proving significantly higher yield for Cans 550 compared to Cans 300.

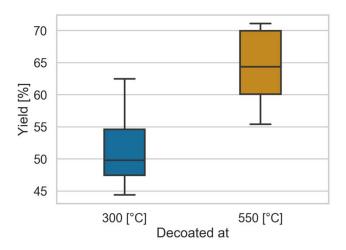


Figure 5: The yield of remelting Cans 300 and Cans 550, with the median values in coloured rectangles representing the quartiles. Average yields and relative dross formations for Cans 300 and Cans 550.

Reduced Pressure Testing

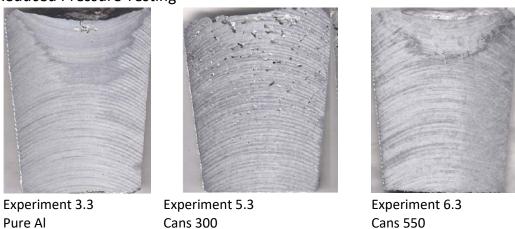


Figure 6: Cut and polished RPT samples from the third Addition in experiments 3, 5 and 6.

In Figure 7 the RPT index, the sums of the length of pores measured on the polished RPT sample, for Pure Al, Cans 300 and Cans 550 are shown. The pure Al has a low RPT index below 50 which indicated as very good metal quality. The Cans 300 ranges from 50 - 200 and while Cans 550 is lower range from 0 - 75. Both the Cans 300 and the Pure Al seems to have declining RPT Indexes as additions increase, possible due to lower relative addition sizes, but a statistically significant trend was not established.

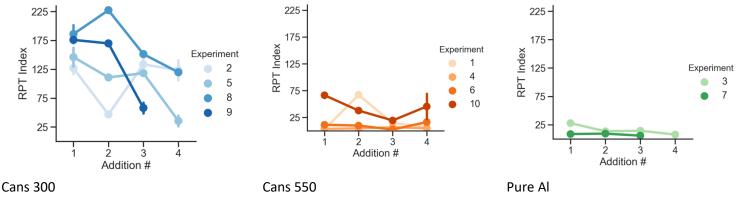


Figure 7: RPT index for each addition in each remelting experiment. Each point is the mean of two measurements accompanied with 95 % confidence lines, which for some points are hidden behind the markers.

Porous Disk Filtration Analysis (PoDFA)

In Figure 8 particle area for mixed oxides are shown in $mm^2 \cdot kg^{-1}$ for the various Pure Al, Cans 300 and Cans 550. Noticeably the readings are quite similar for each material, save for an outlier in Cans 550. Even with this very noticeable outliner, each measurement is well below 0.050 mm²/kg, which is classified as *very light*, the best melt cleanliness in Stanica's classification. [12]. It could be ematnoed that the 8 kg were diluted with 1 kg, but it is believed that this will not change the results.

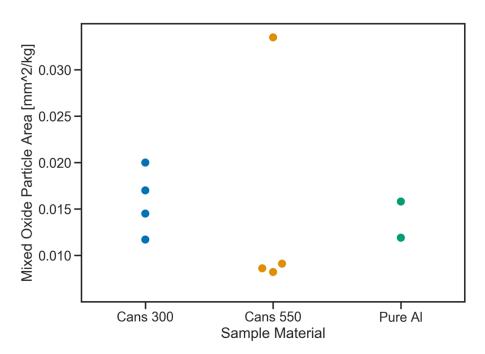


Figure 8: Mixed Oxide Particle area of Cans 300, Cans 550 and Pure Al. Each measurement, including the outlier of Cans 550, is well within Stanica's *very light* classification of melt cleanliness. [12]

Alloy chemistry

In figure 9 below the alloy contents in remelted Cans 300, Cans 550 and pure Al are shown. Remelted Cans has higher contents of alloying elements than remelted pure Al. Notably the concentrations of the alloying elements repressed by the dilution of the Cans sample material with 20 % pure Al, as the in total 8 kg of Cans was melted by submersion in 2 kg of pure Al.

Cans 300 has significantly higher concentrations of Mg and Pb. Due to high variance in measurements differences in concentrations of Sn and Zn were not shown, but Sn is expected to behave similarly to Pb in the sense that its melting point, vapor pressure and formation enthalpy of oxides are similar.

Statistical analysis showed significantly higher concentrations of Magnesium and Lead in Cans 300 compared to Cans 550. The contents were 0.358 % and 0.104 % of magnesium and 11 ppm and 5 ppm of lead in Cans 300 and Cans 550 respectively.

Cans was handled with iron equipment during both decoating and remelting. Some of the iron stems from this equipment, but how much is uncertain.

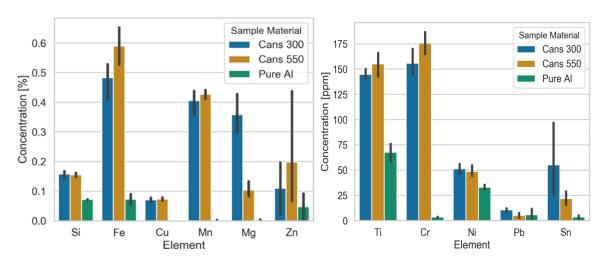


Figure 9: Contents of major (left) and minor (right) alloying elements in each sample material.

Discussion

Due to the low amount of data available the preliminary experiments are used for qualitative considerations. Whilst Cans had a comparable yield to UBC, Foils had a much lower yield, to the extent of being negative. Foils is challenging to decoat due to an organics content over 90 %. The decoated Foils is covered in ash trapped between its many thin layers that must be extracted. With present recycling practices and technology recycling of the aluminium in thin laminates is unfeasible, and substitutes to aluminium should be utilized in such laminates when possible. Whilst the recycling of foils with flux might produce acceptable yields, one would still be burning 20 tonnes of plastic for each metric ton of aluminium recycled.

Remelting of Cans decoated at 550 °C compared to 300 °C resulted in:

- 1. higher yields;
- 2. a lower alloy content;
- 3. better melt cleanliness; and
- 4. a much lower RPT index.

Cans 300 was covered in pyrolysis products. During remelting these would evaporate and combust creating the atmosphere enriched in CO_2 , H_2O and likely CH_4 and CO due to incomplete combustion, which leads to increased metal oxidation and dross production which means reduced yields as discussed previously.

This is the proposed mechanism to explain why remelting of Cans 300 achieved lower yields compared to Cans 550. It could also explain the similar or lower oxide inclusion content of Cans 550, even though Cans 550 was visibly oxidised.

The difference in yields between is more impressive when considering that Cans 300 retained much more of its magnesium throughout the recycling compared to Cans 550. The gas production could explain why Cans 300 retained more of its magnesium throughout the recycling process. An atmosphere of CO_2 has been showed to delay breakaway oxidation of magnesium. Cans 550 however was remelted with much less CO_2 in the atmosphere, and probably had magnesium oxide present before remelting begun, leading to rapid breakaway oxidation of magnesium. If one is able to properly decoat the sample material without producing oxide, this could improve the yield of magnesium during recycling, but losses of magnesium during decoating is probably inevitable considering the high vapour pressure of magnesium at decoating temperature, as seen in table 1.

The lead concentrations in the samples were quite low and measured in ppm. Lead could evaporate to some extent during decoating of Cans 550. The presence of metal droplet proves local melting of aluminium, which in its pure form melts at 660 °C. Furthermore, the material has a very large surface area during decoating compared to the liquid state of remelting, which could help to explain how lead wouldn't evaporate during remelting. It is entirely possible that an unknown reaction mechanism leads to the depletion of lead (and the expected depletion of tin).

The results imply that the equipment used was not fit for decoating due to the uneven temperature distribution during decoating. Ideally the material would be completely decoated with minimal oxide formation. Such a process would be characterised by the highest mass loss during decoating possible, coupled with minimal dross formation during remelting.

Figure 10 ties together the results of the recycling process into the handling of mixed household waste in Norway. $^{\sim}$ 1 % waste might be classified as aluminium waste, and only some of this is recyclable aluminium waste, which can be recycled at a high yield concerning the mass of the aluminium in the recyclable aluminium waste, but the yield per total waste mass is low due to having to remove a large amount of organic matter. In our rather conservative estimates 0.15 % of the mixed household waste, can be extracted as aluminium. If process yield or the fraction of aluminium waste deemed recyclable is increased, this could easily increase up to 0.5 %.

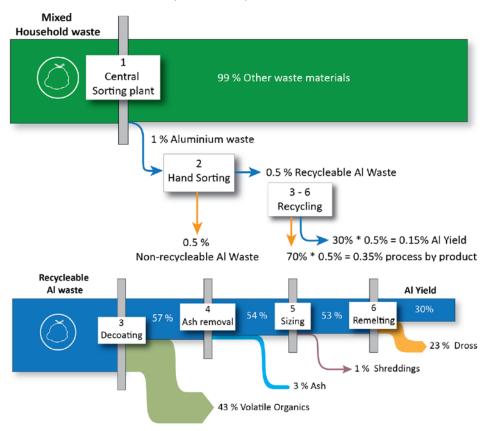


Figure 10: Flowsheet of aluminium waste from a mixed household waste stream, and recyclable aluminium waste (Cans) through the utilized recycling process.

Conclusions

- 1. The Cans sample material can be recycled with acceptable yield and metal quality.
- 2. The Foils sample material is difficult to recycle with acceptable yield and metal quality.
- 3. Decoating Cans at 550 °C compared to 300 °C produced:
 - a. Higher yields;

- b. lower contents of magnesium and lead;
- c. cleaner melt with a lower oxide inclusion area; and
- d. a lower RPT index.
- 4. The content of oxide inclusions in remelted Cans 300 and Cans 550 was classified as very light using cold PoDFA
- 5. Remelted Cans 300 and Cans 550 contains the alloying elements (increasing in order): Cu < Si < Zn < Mg < Mn < Fe.
- 6. The alloy content of elements with noticeable vapour pressures at 550 °C is lower in Cans 550 than in Cans 300.

Future work

In future experiments the variance in pre-treatment and remelting should be kept as small as possible. Sample material should be pre-treated and remelted in the same batches. The same experiments done with Cans should be done with UBC for comparison.

Experiments should be done using the same methodology, but with different varied process parameters, for example: Remelting flux, compaction pressure, ash removal, decoating equipment, shredding of scrap prior to decoating.

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