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Sustainable aluminium recycling of end-of-life products: A joining techniques perspective

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ACCEPTED MANUSCRIPT 1 Word count: 7635 words Sustainable Aluminium Recycling of End-of-Life Products: A 2 **Joining Techniques Perspective** 3 Vi Kie Soo^a*, Jef Peeters^b, Dimos Paraskevas^b, Paul Compston^a, Matthew Doolan^a, 4 Joost R. Duflou^b 5 ^a Research School of Engineering, College of Engineering and Computer Science, The Australian National 6 7 University, Canberra, ACT 2601, Australia 8 ^b KU Leuven, Department of Mechanical Engineering, Celestijnenlaan 300A, B-3001 Heverlee, Belgium 9 * Corresponding author. Tel.: +6-126-125-5941; fax: +6-126-125-0506. E-mail address: vikie.soo@anu.edu.au

10 Abstract

11 The sustainable management of aluminium has become crucial due to the exponential growth in 12 global demand. The transition to a sustainable society with lightweight electric vehicles has led to the increasing use of aluminium in the transportation sector. This has consequently led to the importance 13 14 of aluminium recycling to prevent the valuable material stream going to landfill. In addition, the 15 extraction of primary aluminium has high environmental impact due to the high energy consumption and waste generation in comparison to secondary aluminium processing. Despite being one of the 16 17 most recycled metals, ongoing trends of multi-material designs and the associated joining choices have caused increasing difficulty of separating aluminium with high purity. 18

19 This paper evaluates the types of joining techniques causing impurities in the aluminium streams, and the relationship between particle size reduction and the presence of impurities due to joints 20 21 particularly for end-of-life vehicles. An empirical experiment in a leading European recycling facility 22 was conducted and demonstrated that mechanical fasteners, such as machine screws, socket 23 screws, bolt screws and rivets, are the major types of joining technique causing impurities. Based on the observations from this case study, the characteristics of imperfectly liberated joints are examined. 24 A Life Cycle Assessment (LCA) is also performed to evaluate the environmental impact of recycling 25 26 different aluminium scrap qualities with varying impurity levels. The outcomes are then used to

provide ecodesign guidelines aimed at improving the quality and increase the quantity of recycledaluminium.

29 Keywords

30 Joining technologies; Al recycling; End-of-Life; recycling efficiency; ecodesign; Life Cycle Assessment

31 1 Introduction

32 Aluminium (Al) is used in a variety of products due to its high strength-to-weight ratio, good 33 formability, and high corrosion resistance. The global demand for Al has seen significant growth, 34 leading to the importance of sustainable metal management. The amount of AI used globally has 35 been increasing since 1950, as can be seen in Figure 1, and this trend is projected to continue 36 (Cullen and Allwood, 2013; Martchek, 2006). One of the major concerns is the continuous energy-37 intensive extraction of primary AI to supply for the growing demand worldwide. This activity has 38 contributed significantly to the global carbon dioxide emissions (Norgate et al., 2007). Although Al is 39 one of the highly recycled metals, offering significant energy saving during secondary production, the 40 benefits of AI recycling are influenced by the purity level of scrap sources (Liu and Müller, 2012).



41

42 Figure 1: Amount of primary and recycled Al used globally (International Aluminium Institute, 2017).

43

44 The transportation industry is one of the major consumers of AI worldwide and is responsible for 45 35-40% of the overall AI consumption (Nappi, 2013). In recent years, the focus on producing lightweight vehicles has led to the increasing use of high purity AI in vehicle design to replace 46 47 conventional steels (Goede et al., 2008). Multi-material design concepts have been progressively 48 adopted by vehicle manufacturers due to the emphasis on reducing vehicle mass, thereby lowering 49 the vehicle carbon footprint (Cui et al., 2011; Miller et al., 2000). Al or Al alloys are among the most 50 suitable material category candidates for the manufacture of multi-material car bodies for automotive 51 applications such as Body-in-White (BIW), chassis components, doors closure and outer panels 52 (Carle and Blount, 1999; Hirsch, 2011; Volkswagen Group, 2009).

53 1.1 Joining Trends in Multi-Material Products – An Example using Vehicle Design

54 The increasing use of multi-material products focussing on mass reduction has led to changing 55 joining trends (Groche et al., 2014; Mori et al., 2013; Soo et al., 2015). For example, the material composition in newer vehicle designs has undergone significant transformation particularly with the 56 57 use of light metals, such as AI (Barnes and Pashby, 2000a; Carle and Blount, 1999; Miller et al., 58 2000), and lightweight materials, such as plastics and composites (U.S Department of Energy, 2013). 59 As a result, the choice of joining techniques that are feasible to combine these multi-material combinations is limited (Meschut et al., 2014). Although there are several ongoing developments in 60 more advanced joining technologies, their application in large-scale production is still restricted due to 61 62 the proven design requirements in this risk averse sector (Barnes and Pashby, 2000a, 2000b), and 63 the additional manufacturing costs of installing new equipment (Davies, 2012). Table 1 shows that the Al-intensive vehicle spaceframe structures (Audi A6 and A8) have increased the use of some joining 64 techniques, while reducing others. The large amount of wrought Al used for these luxury vehicles is 65 expected to be adopted also in electric vehicle and mass-optimised vehicle designs. Hatayama et al. 66 67 (2012) have predicted the increasing demand of wrought AI to produce the power-supply box in 68 electric vehicles. The growing AI composition in vehicle designs has led to the increasing use of mechanical joining techniques, such as screws and rivets, and adhesive bonding. In contrast, 69 70 traditional welding techniques (e.g. spot welding and MIG welding) are showing a decreasing trend. 71 The observed joining trends are also supported by the vehicle manufacturers' viewpoint on the

72 development of joining processes used for future large scale vehicle production (Grote and

- 73 Antonsson, 2009).
- 74 Table 1: Joining trends of different vehicle models' spaceframe structure (Adapted from European Aluminium Association,
- 75 2013; Mirdamadi and Korchnak, n.d.).

Joint type	Audi A6	Audi A6		Audi A8	Audi A8		
	2001-2004	2005-2008		1994-2002	2009-present		
Share of point joint	s (%)					0	
Spot welding	91.5	81.0	\downarrow	28.1	7.5	\downarrow	
Stud welding	3.3	6.5	1	0	0		
Clinching	0.9	1.3		10.0	0	\downarrow	
Screw joints	0	0		0	23.6	1	
Rivets	0	5.8	1	61.9	68.9	1	
Share of linear join	ts (%)				\bigcirc		
Laser welding	8.3	3.3	\downarrow	0	8	1	
MIG welding	6	4.3	\downarrow	100	33.3	\downarrow	
Laser brazing	0	3.1	1	0	0		
Adhesive bonding	85.7	89.3	1	0	58.7	1	

76

Perfect material separation during the end-of-life (EoL) phase is not possible in the shredder-77 78 based recycling practices due to the complex product designs and the difficulty in separating different 79 material types from their associated joining techniques (Van Schaik and Reuter, 2007). Castro et al. 80 (2005) and Van Schaik and Reuter (2007) have shown that the increasing complexity in vehicle designs further hinders perfect liberation of dissimilar materials. As a result, lower grades and 81 82 qualities of recyclates will be retrieved due to the presence of impurities that lead to cascade recycling 83 (Paraskevas et al., 2015) and the loss of valuable material streams. This is particularly the case for recycling AI scrap that has more limitations during metallurgical recycling in comparison to other 84 85 metals such as iron and copper (Nakajima et al., 2010). One of the main reasons is the relatively low 86 melting point of AI, which makes it difficult to remove impurities or tramp elements-contaminants affecting the guality of metals that are not added on purpose-during the secondary AI smelting and 87 88 refining processes. The most common strategies used to address this challenge are either dilution 89 using primary AI or down-cycling to lower grade AI alloys that are associated to additional

environmental burden (Castro et al., 2004; Paraskevas et al., 2015). The ability to retrieve high quality
Al with low impurities increases the scrap value for recyclers; however, the extra recycling costs need
to be justified by the volume of different scrap qualities.

93 1.2 Generic Design Guidelines for Joining Choices

94 Most of the design rules specific to joining choices facilitate Design for Disassembly (DfD) for part 95 repair or material reuse that may not cater well for destructive recycling (Ferrão and Amaral, 2006; Güngör, 2006; Kriwet et al., 1995; Reuter, 2011). Screwing, for example, is preferred over adhesive 96 97 bonding to ease the maintenance and repair of highly complex products, as well as part reuse. However, most EoL products are put through a shredding process in industrialised regions. 98 99 Consequently, screws may not be separated well from the base materials, particularly when they are 100 used to combine different material types (Castro et al., 2005; Van Schaik and Reuter, 2007), whereas 101 weak adhesive bonding might be easily released in a shredding process. With the increasing 102 complexity of multi-material structures in products, there is a need to consider the implications of both 103 product design and the choice of joining elements on the quality of recycled materials (Reuter, 2011).

104 There are a few ecodesign guidelines that provide advice on joint selection for destructive 105 recycling, such as the "Ten Golden Rules" (Luttropp and Lagerstedt, 2006) and VDI 2243 design 106 guidelines (VDI 2243, 1993). In spite of that, the details on joint selection are very generic. According 107 to the 'golden rule number 10', joining elements should be minimal, and the use of screws, adhesives, 108 welding, snap fits and geometric locking should be appropriate for different life cycle scenarios. In 109 contrast, VDI 2243 and recycling guidelines by Bras (2005) differentiate the guidelines between 110 disassembly and destructive recycling to assist designers in choosing the most appropriate product 111 design based on the fastening principles.

The choice of joining techniques during the design phase is gaining prominence for the sustainability of high purity metal recycling due to the current recycling practices (Castro et al., 2005; Van Schaik and Reuter, 2007; Worrell and Reuter, 2014). Studies on the limitations of Al recycling are mostly focussed on the efficiency of current sorting and separation processes (Froelich et al., 2007; Gaustad et al., 2012) and the challenges during the metallurgical recycling phase (Nakajima et al., 2010; Paraskevas et al., 2015; Reck and Graedel, 2012). There is a lack of understanding of the

influence of joining choices during initial product design on the quality of recycled AI that is retrievedfrom highly complex products.

120 This work investigates the types of joining techniques causing impurities in the AI streams when 121 advanced recycling technologies are applied. The study is based on an industrial trial carried out in a 122 leading recycler located in Belgium. Impurities due to joints are identified to understand to what extent 123 they are affecting the collected AI streams. The observations are then expanded to assess the correlation between the presence of impurities due to specific joint types, and the different particle 124 125 liberation sizes. Based on the case study data, a Life Cycle Assessment (LCA) is performed to evaluate the environmental impact of recycling different AI scrap qualities. This study assists 126 127 manufacturers and designers to promote closed-loop recycling by mitigating the source of impurities 128 through effective joining technology selection during the initial design stage that caters for current 129 recycling practices. In addition, recyclers and policy-makers can target effective recycling processes and standards to ensure perfectly liberated joints for high purity AI to minimise the loss of valuable 130 131 material streams.

132 2 Materials and Methods

This study analyses the cause of impurities present in the different Al output fractions sampled from a Belgium recycling facility. Section 2.1-2.3 describe the Al recycling processes and sampling procedures. To assess to what extent the impurities are affecting the environmental impact of recycling different Al scrap qualities, the LCA method is used, as detailed in Section 2.4.

137 2.1 Recovery of Aluminium in Belgium

The types of scrap sources as considered in the studied Belgian recycling facility, one of the leading recyclers in Europe, are shown in Table 2. The high content of Al in different scrap sources has made it one of the most intensely recycled metals besides steel. Most of the Al scrap is contributed by the end-of-life vehicle (ELV) and household waste streams. The Al content in the ELV and household waste accounts for 4.9wt.% (Muchová and Eder, 2010; RDC Environment, 2015) and 4.7wt.% (Muchová and Eder, 2010) respectively. The Al content in demolition and building scrap is relatively low, less than 1wt.% (Muchová and Eder, 2010).

145 Table 2: The sources of the AI containing scrap as recycled in the Belgian recycling facility.

Scrap sources	Relative share of total scrap stream (%)
Depolluted vehicle hulks (ELV)	30
Demolition scrap	30
Household waste	20
Building scrap	20

146

The material process flow specific to AI is shown in Figure 2. The processes involved in AI recycling can be categorised into three main clusters: AI sorting, refinement of sorted aluminium, and particle size sorting. For AI sorting, density separation is the first step to retrieve AI from the mixture of scrap. Subsequently, other major processes, such as eddy current separator, optical separator, and head pulley magnet, are used to further separate AI from other material types.





153 Figure 2: AI material flow in the Belgian recycling facility.

154

The sorting process targeting AI begins with the density separation after the shredding and magnetic separation. Density separation sorts different materials based on their material densities. It typically starts with separating lighter material fractions (e.g. plastics, foam, rubber, etc.) and is followed by the sorting of materials with higher density. Through density separation at 3kg/l, AI alloys float and can be separated from materials with higher density, such as copper, zinc, and other heavy metals, that sink to the bottom. In some other recycling facilities, AI retrieval through eddy current separation is carried out (Gaustad et al., 2012).

An air classifier is used to remove fine shredder residues targeted for energy recovery before the density separation. This allows the fine mixture of dust, metals, glass, and polymers to be removed before the first density separation for lighter material fraction. Other separation techniques, such as sievers, are also used. The material flow is sorted to different particle sizes based on the siever sizes used at various screening stages. This is a common practice in the recycling industry in Europe to segregate different material grades based on the particle sizes (Cui and Forssberg, 2003).

168 An eddy current separator, optical separator and head pulley magnet are used to further sort 169 unwanted materials that are still present in the Al flow. The remaining cable wires that did not sink 170 during earlier density separation are further sorted using the eddy current separator. Through this 171 process, an electrical current is induced within the conductive metal flow, and all metals are repelled through the rotor that produces an external magnetic field. Since AI and copper have a different 172 173 conductivity, and thus produce varying eddy currents, they are ejected to different distances from the 174 rotor. An optical separator is then utilised to further sort the commonly green coloured printed wiring 175 board (PWB) from the grey coloured aluminium. To further remove small particles with ferrous content 176 from the Al flow, a very strong head pulley magnet with a deeper magnetic field is used.

177 2.2 Sampling Method

The different AI fraction categories recovered from the facility are shown in Table 3. These categories were chosen for sampling to understand the effect of particle sizes on the purity level of various AI fractions, and the extent of impurities due to joints in the different particle sizes. Sampling was also carried out for AI with high Fe content. The collection of a minimum of 10 samples from each AI fraction was performed in accordance with the field sampling guidance for shredded scrap by the

United States Environmental Protection Agency (USEPA, 1993). The field sampling guidance provides information on different sampling methods, estimated sample size, and the statistical analysis methods to accurately approximate the impurity level of different AI fractions. These guidelines were based on previous case studies carried out at different shredder sites.

187 Table 3: Amount of Al samples taken from each category, and the generated annual amount in the Belgian recycling facility.

Category	egory Particle Number		Mass range of	Overall	Annual
	size class	of	each sample (kg)	sample mass	amount
	(mm)	samples		(kg)	(ton)
Al with high Fe content	12-120	10	2.685-3.737	32.689	644
AI fraction	40-120	20	2.290-3.896	61.363	6132
AI fraction	12-40	10	1.506-2.408	19.210	4147
Al fraction	4-12	10	1.494-1.947	16.662	1114

188

There are different field sampling methods for shredded metal scraps on-site based on the 189 guidelines by USEPA. Stockpile sampling, as explained in Figure 3, was chosen in this case study to 190 obtain a more representative sample of the normal shredder output (USEPA, 1993). Al samples were 191 192 taken from the AI stockpile warehouse where different qualities and particle sizes were stored 193 separately. The bucket used to collect the samples has a diameter of 27.5cm with a height of 22.5cm. 194 Each sample taken only filled up half the bucket. First, Al samples were collected at the edge of pile (location 1) at notch 1 and notch 2. The two notches were then dug to equal depth with the help of a 195 196 front-loader truck. Finally, samples were gathered at locations 2 to 5 for notch 1 and 2. In total, there 197 were 10 buckets of samples collected for each targeted Al output stream. 20 samples were taken only 198 for the AI fraction 40-120mm to ensure a good representation of the stockpile, since it is the largest 199 fraction produced in the facility.



Figure 3: Stockpile sampling of different AI fractions (a) Sampling location for each AI stockpile; (b) Location of notches made
for each AI stockpile to carry out sampling.

202

203 2.3 Sample Analysis Procedures

The AI with high Fe content fraction was sieved through a 40mm mesh siever to separate particles to two particle size categories: 12-40mm and 40-120mm. This step was carried out to allow comparability with the observations made for the AI fractions of similar particle size classes.

Each particle was weighed and hand-sorted according to the different liberation classifications, as shown in Figure 4 and as follows.

209	•	Liberated AI samples consisting of AI only (Figure 4a).
210	•	Liberated impurities were particles consisting of a single material type other than Al
211		(Figure 4b).
212	٠	Unliberated impurities were particles consisting of material combinations other than Al
213		(Figure 4b).
214	•	Unliberated AI samples were particles consisting of AI that was still attached to other
215		material types without the presence of a joint (Figure 4c).
216	•	Unliberated AI samples due to joint were particles consisting of AI that was still attached
217		to other material types with the presence of a joint (Figure 4d).



Figure 4: Examples of liberation classification for particles end up in the AI stream. (a) Liberated AI samples (AI particles only);
(b) Liberated/unliberated impurities (liberated glass and unliberated Cu-Fe particles); (c) Unliberated AI sample not due to joint
(PWB inserted in AI particle); (d) Unliberated AI sample due to joint (screw and bolt attached to AI particle).

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Unliberated particles were further separated into their individual materials. The mass of each material was recorded. For cases where further material separation was not possible due to entanglement or rust, the mass of individual materials was calculated using their volumes and average material densities: AI (2745kg/m³), steel (7825kg/m³), copper (8595kg/m³), plastic (1253kg/m³), rubber (1270kg/m³), and foam (255kg/m³) (Callister and Rethwisch, 2013; Gaustad et al., 2012). The types and characteristics of joints causing impurities were observed, and the range of joint sizes, joint material liberation, and the number of rusty joints were recorded quantitatively.

229 2.4 Er

Environmental Impact Assessment

To evaluate the environmental impacts associated with the quality of different AI scrap fractions collected from the case study, LCA was carried out to assess the dilution and quality losses in remelting the scrap to be reused as AI 6061 alloy (AA6061). During remelting, dilution losses occur due to the need to dilute the residual element concentration (e.g. Fe) with primary AI. To avoid quality losses, alloying elements (e.g. Si and Cu) are added (Paraskevas et al., 2015). The environmental impact assessment only takes into consideration the secondary AI processing of the defined system boundary shown in Figure 5. The wrought AA6061 was chosen as the target secondary alloy since it

is widely used in automotive applications and thus, likely to be close to the average composition of the Al scrap retrieved from ELV. To compare the environmental impact of smelting different Al scrap, the functional unit is defined as Al recycling to produce 1 tonne of AA6061. The calculations for the required primary Al for dilution purposes and the additional alloying elements are attached in Appendix A. The credits for subsequent recycling of by-products, such as dross and salt slag, were also taken into consideration.



244 Figure 5: System boundary and functional unit of secondary AI processing for different AI scrap fractions.

245

246 GaBi software was used to model all the processes and resources involved during the secondary 247 Al processing. Electricity generation was modelled based on the average electricity consumption mix in Europe. The life cycle inventories were obtained from GaBi Professional database v6.115 and a 248 249 previous comprehensive report from the Aluminium Association (The Aluminium Association, 2013), as detailed in Table 4. The environmental performance was calculated based on the midpoint 250 251 categories of the International Reference Life Cycle Data System (ILCD recommendations v1.09). 252 These recommendations were based on the ILCD handbook in accordance with the ISO 14040 series 253 (European Commission et al., 2010; ISO, 2006). Following this method, the midpoint results were 254 normalised to person-equivalent (PE) units-the environmental impact caused by an average 255 European annually. An equal weighting was applied for the midpoint impact categories, and the

normalised PE scores for all midpoint categories were added to allow comparison of the overall environmental performance for different AI scrap fractions to achieve 1 tonne of AA6061. The single impact score for climate change (include biogenic carbon) and resource depletion, mineral, fossils and renewables impact are provided in Table 4 due to their significant contribution to the environmental performance.

Table 4: The life cycle inventory data sources for materials and recycling processes, and the respective impact score for climatechange (include biogenic carbon) and resource depletion, mineral, fossils and renewables.

Process	Climate	Mineral and	Source	Description
	change	fossil depletion		
	(PE/ton)	(PE/ton)		
Al scrap	6.01E-03	3.62E-04	(The Aluminium	The dataset includes scrap
preprocessing			Association,	collection, separation, cleaning,
			2013)	and preprocessing.
Al scrap	2.59E-01	1.15E-02	(The Aluminium	The dataset includes remelting,
remelting			Association,	refining, alloying, and casting of
			2013)	secondary Al. Dross and salt
				slag recycling are included.
Primary Al	1.08E+00	2.84E-01	GaBi	The dataset includes cradle to
ingot			Professional	gate inventory for primary Al
			Database	ingot production in Europe.
			v6.115	
Primary Cu	4.04E-01	3.56E+02	GaBi	The dataset includes cradle to
			Professional	gate inventory for primary Cu
			Database	(99.999%) in Germany.
			v6.115	
Primary Si	5.45E-01	4.77E-02	GaBi	The dataset includes cradle to
			Professional	gate inventory for primary Si
			Database	(99%) in global context. The
			v6.115	chemical composition is
				approximated based on Si-2202
				(BAIDAO, 2007; SINOGU,
				2016).

263

264 3 Results

The liberation categories of the collected Al samples from different fractions were studied. The average Al purity of each fraction was determined. The presence of impurities due to joints was further analysed, and the types of joining techniques causing impurities were characterised.

268 3.1 Al Sample Analysis

269 The mass distribution of particles in the different liberation categories is shown in Figure 6. 270 Liberated and unliberated impurities were mainly caused by separation errors during the recycling 271 processes, and can be characterised as fine particles (<4mm); materials with similar density range to Al; small and longitudinal heavy metal particles; and materials with density less than Al (<2kg/l). The 272 273 types of impurities consisted of ferrosilicon fines, glass, PWB, Cu, Fe, wires, plastics, and other light fraction of non-metals. Ferrosilicon fines are an example of fine particles easily trapped in Al samples 274 275 during the density separation. Glass and PWB have a density range of 2.47-2.54kg/l (Malone and 276 Dolter, 2008) and 1.5-2.89kg/I (Bizzo et al., 2014; Zhang and Forssberg, 1997) respectively that can 277 be similar to AI density. Small heavy metal particles, such as Cu, Fe and wires, with thin and long 278 shapes caused them to be entangled between AI particles during the density separation. Plastics, rubber, fabric, fibrous materials and foam are examples of impurities that were not well separated 279 280 through density separation at earlier stages. Unliberated AI samples both with or without the presence 281 of joint have higher AI content in the particles by mass and therefore, they were more likely to end up 282 in the AI streams.





285

283

From Figure 6, the total mass of unliberated AI samples both with and without joint is showing an increasing trend when the particle sizes are larger. This observation is valid for both the AI fraction and AI with high steel content fraction. To understand the purity level of AI samples for different particle sizes, the mass fraction of impurities was calculated and the result is shown in Table 5.

290 Table 5: Al purity for different Al fractions with 95% confidence interval.

Category	Particle size class (mm)	Al purity (wt.%)
Al with high Fe content fraction	40-120	82.07 ± 3.86
Al with high Fe content fraction	12-40	80.75 ± 3.38
Al fraction	40-120	98.66 ± 0.58
Al fraction	12-40	99.57 ± 0.29
Al fraction	4-12	98.11 ± 0.58

291

In general, the quality of recycled AI can be separated into two classes: AI purity more than 98%,

and AI purity less than 83%. AI purity less than 83% consisted of AI with high steel content fractions

that were separated through a strong head pulley magnet as the final separation process in the recycling facility.

Based on the analysis of the shredded samples, smaller particle sizes do not indicate higher Al purity. Al with high Fe content fraction (40-120mm), and Al fraction (12-40mm) have higher Al purity values in their respective categories. The geometry, joint size, and material types of the combined parts also affect the purity levels of Al fractions in different particle sizes. For instance, when a large number of small steel screw fasteners (e.g. steel screw with diameter and length of 2mm and 4mm respectively) are used, the likelihood of Fe impurities due to screw fasteners present in the Al fraction in smaller particle sizes is quite high with respect to mass.

303 The material types of impurities were identified to understand the extent of contamination in the AI 304 samples. Some of the impurity types can be removed easily during the secondary Al production 305 whereas others, such as Fe, require a dilution process using primary Al. As seen in Table 6, the types 306 of impurities are Fe, Cu, organic and inorganic. It can be observed that the smaller particle size 307 fraction, 4-12mm has a higher impurity level than the 12-40mm fraction due to the material types and 308 the physical characteristics of impurities. These impurities are largely contributed by ferrosilicon fines 309 (consisting of ferrous and silicon), thin and long-shaped wires (consisting of Cu and plastics), small 310 pieces of shattered glass (silicon) and plastics that typically have small dimensions or high brittleness. One of the most undesired tramp elements during AI recycling are Fe impurities (Cho et al., 2015; 311 312 Paraskevas et al., 2015) due to their detrimental effect on the mechanical properties of Al alloys 313 (Belov et al., 2002). Therefore, this case study focussed on the source of Fe impurities in unliberated 314 samples due to joints to understand the impact of joining choices on the purity level of recycled AI.

315 Table 6: Types of impurities present in the AI output streams in the Belgian recycling facility.

Category		Particle size	Average mass percentage (wt.%)					
	Y.		Fe Cu		Organic	Inorganic		
			impurities	impurities	impurities	impurities		
Al with high	Fe fraction	40-120	11.32	0.27	5.82	0.42		
Al with high	Fe fraction	12-40	9.82	1.38	6.40	1.56		
Al fraction		40-120	0.36	0.25	0.71	0.05		
Al fraction		12-40	0.03	0.13	0.23	0.06		
Al fraction		4-12	0.14	0.26	0.96	0.43		

316 3.2 Observations on the Joint Type Causing Impurities

From the collected AI samples, it was observed that mechanical fastening and adhesive bonding were the two main types of joining techniques causing impurities. The amount of unliberated AI samples due to adhesive bonding was extremely small. They were mostly combinations of AI and lower density materials, such as AI-plastic and AI-foam particles, using lap joint. Lower density materials assisted in breakage during the shredding process due to centrifugal force, and hence, were less likely to cause impurities in the AI samples.

In contrast, mechanical fasteners were the major type of joining method contributing to the presence of Fe impurities in the AI stream, since they are typically made of steel. They were further classified to understand the different types of mechanical fasteners, and how their characteristics contributed to the presence of impurities. Figure 7 shows the various types of mechanical fasteners that were observed in the unliberated AI samples due to joints.



Figure 7: Classification of different mechanical fastening joining methods (Bolt Depot, 2013). (a) Machine screw; (b) Bolt screw;
(c) Socket screw; (d) Rivet; (e) Pin; (f) Steel clip.

330

331 3.2.1 Al with High Steel Content Fraction (12-120mm)

In Figure 8, it can be observed that the likelihood of Fe impurities due to separation errors decreases for larger particle sizes in Al fraction. The Fe impurities observed in the Al with high Fe content fraction (12-40mm) were mostly small in size due to the inability of the head pulley magnet to pick up small Fe content. There were no Fe impurities due to separation errors observed in the larger particle sizes. Fe impurities that were larger in size have higher likelihood of being separated by the

magnetic separator after the shredding process. On the other hand, impurities due to imperfect material liberation were largely caused by structural design, such as enclosures (parts surrounded by different material types) and entanglement (parts that were twisted together or caught in), after the shredding process. Therefore, the likelihood of Fe impurities due to imperfect material liberation increases for larger particle sizes in the Al fraction.





(a) Al with high steel content fraction (12-40mm) without Fe impurities due to unliberated rivets and pins

Impurities due to separation error

- Impurities due to unliberated joint-Machine screw
- Impurities due to unliberated joint-Bolt screw
- Impurities due to unliberated joint-Steel clip

(b) Al with high steel content fraction (40-120mm) without Fe

impurities due to separation errors

Impurities due to imperfect material liberation Impurities due to unliberated joint-Socket screw Impurities due to unliberated joint-Rivet Impurities due to unliberated joint-Pin

342 Figure 8: Fe impurities present in the AI with high steel content fraction with 95% confidence intervals.

343

344 There were a variety of mechanical fastener types causing Fe impurities in the AI with high Fe 345 content fractions. Fe impurities observed in smaller particle sizes were caused by unliberated 346 machine screws, socket screws, bolt screws, rivets, and steel clips. No pins were observed for this fraction due to the smoother joining surface that allowed them to be well liberated when shredded to 347 348 smaller particle sizes. The types of mechanical fasteners causing Fe Impurities in the larger particle 349 sizes were machine screws, socket screws, bolt screws, rivets, steel clips and pin. For both fractions, 350 machine screws were more likely to cause impurities when compared to other mechanical fastener 351 types.

88

The types of mechanical fasteners causing impurities were further characterised through 352 353 observation of their physical attributes, as shown in Table 7. The percentages are with respect to the 354 total number of units for each joint type. It is observed that the number of mechanical fasteners in the 355 larger particle sizes were higher compared to the smaller particle sizes except for machine screws 356 and steel clips. Moreover, the fraction with larger particle sizes has a wider range of joint sizes when 357 compared to smaller particle sizes. In spite of that, the number of joint sizes with diameter and length more than 6mm and 10mm respectively (large joint sizes) is similar for both particle size classes. 358 359 Partial liberation joints, those with more than 50 wt.% of the joint material liberated, were more likely for threaded fasteners such as machine screws and bolt screws. In most cases, the fasteners' head 360 361 was liberated due to protrusion. Rusty threaded fasteners were also more likely to cause impurities in 362 the AI samples.

363	Table 7: Characteristics of joint causing Fe impurities in AI with high Fe content fraction	'n

Joint types	Total	Joint size (mm)		Large joint Partial		Rust (%)
	(unit)	Diameter	Length	size (%)	liberation (%)	
12-40mm						
Machine screw	101	2-10	3-30	12	9	86
Socket screw	11	4-7	9-36	27	0	64
Bolt screw	16	4-10	8-50	56	0	94
Rivet	13	4-5	3-13	0	0	46
Steel clip	19	2-3	10	0	0	0
40-120mm						
Machine screw	94	2-12	2-30	12	4	76
Socket screw	20	3-9	10-60	30	0	85
Bolt screw	39	3-14	7-125	52	8	76
Rivet	48	5-6	3-50	1	0	48
Steel clip	2	2-3	12	0	0	0
Pin	1		10	0	0	0

364

Al Fraction (4-120mm) 365 3.2.2

Similar to AI samples with high Fe content, the likelihood of Fe impurities due to separation errors 366 decreases for larger particles sizes in the Al fraction, as seen in Figure 9, since they can be easily 367 368 sorted through magnetic separation. In contrast, impurities due to imperfect material liberation could

369 potentially be higher for larger particle sizes, although they were not observed in the AI fraction (12-

370 40mm).





(a) Al fraction (4-12mm) with Fe impurities due to separation errors, imperfect material liberation, and unliberated machine screws

(b) Al fraction (12-40mm) with Fe impurities due to separation

errors and unliberated machine screws



of unliberated joint types

Impurities due to separation error

- Impurities due to unliberated joint-Machine screw
- Impurities due to unliberated joint-Bolt screw
- Impurities due to imperfect material liberation
- Impurities due to unliberated joint-Socket screw
- Impurities due to unliberated joint-Rivet
- 371 Figure 9: Fe impurities present in the AI fraction with 95% confidence intervals.

372

The likelihood of Fe impurities due to mechanically fastened joints in the Al fraction is higher for larger particle sizes. There was more variety of mechanical fastener types that contribute to the Fe

impurities in the Al fraction (40-120mm). Machine screws were the only type of joint causing impurities
in the smaller particle sizes, whereas machine screws, socket screws, bolt screws and rivets were
observed in Al fraction (40-120mm). Despite the use of a strong head pulley magnet to remove small
Fe content, machine screws contaminating the different Al fractions were still present.

379 Table 8 shows the attributes of mechanical fasteners causing Fe impurities in the different AI 380 fractions. The number of machine screws observed in Al fraction (40-120mm) was larger compared to 381 the fraction containing the smaller particle sizes. However, there was still a small number of machine 382 screws present in this smaller particle size fraction. This was due to the lower magnetic force experienced by small screws located at enclosed spots despite the use of a strong head pulley 383 384 magnet. In contrast, the presence of mechanical fasteners other than machine screws (socket screws, 385 bolt screws, and rivets) was only seen in Al fraction (40-120mm). Socket screws and bolt screws have 386 a more protruded head compared to machine screws that facilitate liberation during the shredding process. On the other hand, rivets have a smooth surface that allows them to be easily set free when 387 388 shredded into smaller particle sizes. The likelihood of impurities due to larger joint sizes or of partial 389 liberation is higher for larger particle sizes particularly for the machine screw fastener type.

Joint types	Total	Joint size (mm)		Large joint	Partial	Rust (%)
	(unit)	Diameter	Length	size (%)	liberation (%)	
4-12mm			Y			
Machine screw	2	3-4	8	0	0	100
12-40mm	~					
Machine screw	1	5	20	0	0	100
40-120mm	\bigcirc					
Machine screw	17	3-8	5-25	18	12	50
Socket screw	1	4	18	0	0	0
Bolt screw	2	4	11-12	0	0	50
Rivet	2	5	7	0	0	0

390 Table 8: Characteristics of joint causing Fe impurities in Al fractions.

391

392 4 Environmental Impact Assessment Results

393 From the LCA results presented in Figure 10, the total environmental impact for Al with high Fe scrap fractions (both particle sizes) has increased by at least 28 times in comparison to the Al scrap 394 395 fractions (4-12mm, 12-40mm, and 40-120mm). This is caused by the higher concentration of Fe, that can be considered as impurities rather than useful alloying elements, and the addition of Si and Cu 396 397 alloying elements to produce AA6061. The contribution of different midpoint impact categories for the 398 different AI scrap fractions to produce 1 tonne of AA6061 is provided in Appendix A. The use of primary AI as dilution agent is the major contributor to the environmental impact for AI scrap with high 399 400 Fe content with an impact share of at least 92%, as supported in other studies (Amini et al., 2007; 401 Paraskevas et al., 2015). To achieve higher purity wrought Al alloy, a substantial amount of primary Al 402 is required for the dilution of these streams, and alloying elements are added to meet the compositional limits. This results in scrap underutilisation since only 3-6wt.% of the produced AA6061 403 404 consists of recycled AI scrap. The use of primary AI for dilution can be minimised by using other high 405 purity scrap streams and optimised Al scrap blending (Paraskevas et al., 2015).



406

407 Figure 10: Total environmental impact and the share of recycling, dilution and quality losses for different AI scrap fractions to408 achieve 1 tonne of AA6061.

409 A sensitivity analysis was carried out to assess the influence of varying element values for 410 different AI scrap fractions. The range of values for Fe, Si and Cu elements are shown in Table 9. As 411 can be seen from Figure 10, the total environmental impact is sensitive to the range of values for Fe, 412 Si and Cu in different Al scrap fractions. It is shown that the margin of error for the total environmental 413 impact can be up to ± 0.5 person equivalent per tonne. However, the trend of the total environmental 414 impact for the range of element values is largely unaffected. The total environmental impact for AI with 415 high Fe scrap fractions is greatly influenced by the dilution losses. In contrast, the total environmental impact for AI scrap fractions is largely contributed by recycling process and quality losses since no 416 417 dilution losses occur. Despite the sensitivity to the range of Fe, Si and Cu values, the negative impact 418 of the AI scrap fractions is insignificant compared to AI scrap with high Fe content.

419 Table 9: The range of values for Fe, Si and Cu present in the different AI scrap fractions with 95% confidence interval.

Category	Particle sizes (mm)	Fe (wt.%)		Si (wt.%)		Cu (wt.%)	
		min	max	min	max	min	max
Al with high Fe fraction	40-120	9.95	12.69	0.00	0.94	0.00	0.46
Al with high Fe fraction	12-40	7.12	12.53	0.00	3.51	0.51	2.24
AI fraction	40-120	0.03	0.68	0.00	0.11	0.06	0.46
AI fraction	12-40	0.00	0.06	0.03	0.09	0.00	0.28
Al fraction	4-12	0.07	0.20	0.27	0.54	0.14	0.38

420

421 As can be seen from Figure 11, about 70% of the total impact share of dilution losses for Al scrap 422 with high Fe content is caused by unliberated joints. Dilution losses due to material separation errors 423 can only be observed for AI with high Fe scrap in smaller particle sizes due to the presence of silicon from the shattered glass. The environmental evaluation based on the case study data shows that the 424 425 dilution and quality loss impacts are tightly-linked to the quality or purity level of the recovered AI streams resulting from material liberation. The high Fe content due to improper material liberation has 426 427 become a limiting factor for the recyclability of the AI streams. It is worth noting that the environmental 428 performance may vary according to the efficiency of recycling technologies used in different countries.



430 Figure 11: The impact share of dilution losses (primary Al addition) and the environmental impact (person equivalent per tonne)431 due to liberation categories.

432 5 Discussion

The annual AI output streams in the Belgian recycling facility are shown in Table 10. The material 433 434 composition in each AI stream is estimated from the performed sampling, and subjected to the 435 variation based on the 95% confidence interval. Despite the large variance for certain tramp elements 436 in different AI output streams, the impact on the environmental performance is insignificant. AI 437 fractions (12-40mm) and (40-120mm) are the two largest fractions with high AI purity levels of 99.57% 438 and 98.66% respectively, whereas the AI with high Fe content fraction has the lowest annual amount with low AI purity level of 81.28% (combination of both particle sizes). With the increasing complexity 439 of multi-material designs, particularly in the automotive sector which is one of the largest consumers 440 441 of AI, it is projected that the AI with high Fe content fraction will be growing and thus, lead to the 442 reduction of the AI fraction with higher purity (Soo et al., 2015, 2016).

444 extrapolation of sampling results.

Material type	Al with high Fe		Al fraction		Al fractio	Al fraction		n
	content	fraction	(40-120m	m)	(12-40mm	n)	(4-12mm))
	(12-120	mm)						
	ton	wt. %	ton	wt. %	ton	wt. %	ton	wt. %
AI	523.46	81.28	6048.50	98.64	4128.60	99.56	1093.00	98.11
PWB	2.09	0.32	3.14	0.05	1.45	0.03	6.04	0.54
Wire	4.21	0.65	18.80	0.31	2.41	0.06	2.72	0.24
Cu	0.62	0.10	7.60	0.12	4.24	0.10	0.61	0.06
Plastic/composite	12.13	1.88	19.44	0.32	3.87	0.09	3.57	0.32
Rubber	18.26	2.84	6.71	0.11	2.67	0.06	0.20	0.02
Steel	75.82	11.77	21.39	0.35	0.73	0.02	1.29	0.12
Foam	0.57	0.09	0.46	0.01	0.21	0.01	0	0
Fabric	2.19	0.34	2.16	0.04	0	0	0	0
Synthetic leather	0.28	0.04	0	0	0	0	0	0
Glass	0	0	0	0	0	0	5.06	0.45
Fibrous material	2.66	0.41	0	0.41	0	0	0.40	0.04
Ferrosilicon fine	1.70	0.26	3.86	0.06	2.80	0.07	1.16	0.10
TOTAL:	644	100	6132	100	4147	100	1114	100

445

446 5.1 Observational Study Outcomes

From the analysed samples, most of the Fe impurities were due to unliberated joints particularly 447 for AI with high Fe fractions, and AI particles of larger sizes, as seen in Table 11. Particles with 448 449 unliberated joints in the AI with high Fe fractions have contributed at least by 69% to the total Fe 450 impurities. When the particle sizes for different AI fractions decrease, the total Fe impurities due to 451 unliberated joints decrease by at least 33%. Therefore, smaller particle sizes can assist in reducing Fe impurities due to unliberated joints. However, the proportion of Fe impurities due to separation errors 452 or imperfect material liberation is higher for Al fractions with smaller particle sizes. The presence of 453 454 these impurities is strongly influenced by the material structural design, joint size used, and the efficiency of the recycling processes in sorting small to fine particles. Thus, additional loops in Fe 455 impurity removal or adjustment of the installation with strong magnets could assist in reducing 456 457 material separation errors for smaller particle sizes.

⁴⁴³ Table 10: Estimated material composition for the Belgian recycling facility's annual AI output streams based on the

Category	Particle size class (mm)	Total Fe impurities (wt.%)		
		Separation	Imperfect material liberation	Unliberated joint
		error		
Al with high Fe fraction	40-120	0	3.52	7.88
Al with high Fe fraction	12-40	1.41	1.47	6.94
Al fraction	40-120	0.04	0.08	0.24
Al fraction	12-40	0.03	0	0.01
Al fraction	4-12	0.06	0.07	0.01

458 Table 11: The proportion of Fe impurities due to separation error, imperfect material liberation and unliberated joint.

459

460 5.2 Recommendations to Improve AI Scrap Quality

461 Based on the case study observations, the suggestions to improve the quality of AI recycling from 462 ecodesign (materials and connections) and recycling process perspectives are as follows.

463 Ecodesign

464	•	Encourage the use of low cost disassembly embedded design, such as the use of active
465		fasteners (connections that can be unfastened simultaneously through a specific trigger
466		or a combination of triggers), to maximise the material/part reuse without compromising
467		the product use phase (Duflou et al., 2008; Peeters et al., 2015; Peeters et al., 2017).

- Encourage the use of mechanical fasteners with greater protrusion and smoother joining
 surface, such as socket screws, rivets and pins, to ease particle liberation.
- Avoid the use of machine screws. Otherwise, minimise the total number and sizes of
 joints particularly for machine screws to reduce the mass of Fe impurities.

472 Recycling processes

- Encourage shredding of particles into smaller sizes to decrease the presence of
 impurities due to mechanical fastening joints and imperfect liberation.
- Encourage the use of strong head pulley magnets to further sort small Fe content
 particularly for Al particles that still contain smaller size mechanical fastening joints. This
 process is not commonly used in recycling facilities.

Encourage the use of more efficient sorting processes for PWB, Cu and wires to refine Al
scrap after density separation, such as wet shaker tables (Jordão et al., 2016), kinetic
gravity separators (Rem, 2009), and nail roll separators (Fabrizi et al., 2003).

The feasibility of the proposed recommendations can be influenced by other factors, such as the economic aspects and legislative boundaries (Soo et al., 2017). Recycling of high purity materials can be affected by the additional recycling costs, the profit of the end products, or the generated mass or volume of different quality scrap fractions. In addition, a governmental role can also be of importance through the imposing of a minimum purity level required for outputs from recycling activities.

486 6 Conclusions and Future Work

Despite the rigorous recycling processes used in the Belgian recycling facility, unliberated joints are one of the major contributors to the presence of Fe impurities in the Al output streams, particularly for Al with high steel content fractions. The Fe content level highly influences the recyclability of Al scrap; thus, the environmental impact of dilution and quality losses during Al recycling needs to be integrated into LCA for better-informed decisions towards closed-loop recycling.

The main type of joining techniques causing impurities in the AI streams are mechanical fasteners, such as machine screws, socket screws, bolt screws and rivets, which are commonly used for assembling AI with other materials. Although adhesive bonding was also observed to cause impurities in the AI particles, these were relatively small and almost negligible when compared to the effects of mechanical fastening joints.

497 Based on the observations of the collected samples, machine screws were the major type of mechanical fasteners causing Fe impurities in different Al fractions due to their joint characteristics. 498 499 This was consistently observed for various particle sizes. Machine screws are normally less protruded 500 compared to other mechanical fasteners, such as bolt screw, and socket screw. A higher level of 501 protrusion eases liberation during the shredding process. In addition, machine screws that were 502 smaller in size, and corroded due to moisture will make particle liberation more challenging. There 503 were also cases of partial liberation due to the threaded structure that further hindered full material 504 liberation.

505 Unliberated AI samples due to the presence of joints are less likely for smaller shredder output 506 fractions with respect to the total mass of particles. It was found that smaller particle sizes ease 507 liberation of Fe impurities from the joints. However, when considering the AI purity level for different 508 particle sizes, they do not indicate a higher purity level for smaller particle sizes. This was largely 509 caused by the increasing proportion of Fe impurities due to separation errors and imperfect material 510 liberation. Although sorting of Al scrap into different fractions is proven to be effective in obtaining high guality Al in most European countries, it is important to understand the guality of recycled Al scrap in 511 512 high consumption countries, such as in China (RBC Capital Markets, 2015), from a global 513 perspective.

It can be concluded that the choice of joining techniques during the design phase has a significant impact on the environmental performance during the EoL phase. The share of unliberated joints causing the environmental impact due to dilution losses in the Al scrap with high Fe content is the highest compared to material separation errors and imperfect material liberation. Dilution losses cause a significant environmental impact and reduce the avoided environmental impact during Al recycling.

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524 Appendix A. Supplementary Data

525 The supplementary data related to this article is as attached.

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Highlights

- Identified the types of joining techniques causing impurities in the AI streams.
- Characterised the joints causing Fe impurities in the AI streams.
- Recycling efficiency of AI was measured through an industrial trial in Europe.
- Identified the linkage between particle sizes and impurities due to joints.
- Assessed the environmental impact of dilution and quality losses due to joints.

Chillip Marker