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Investigation on Resource-Efficient Aluminium Recycling – A State of the Art Review

CHRISTINA WINDMARK^{a,1}, LUCIA LATTANZI^b, ANDRÉ MÅNBERGER^c and ANDERS E. W. JARFORS^b

^a Production and Materials Engineering, Lund University ^b Materials and Manufacturing, School of Engineering, Jönköping University ^c Environmental and Energy Systems Studies, Lund University

Abstract. Recycling is an important area to improve to reduce negative impact on the environment. With increased material recovery, less virgin materials are needed to provide the same benefits for the society. Aluminium is an important metal in the efforts to reduce negative climate impact. Demand for wrought aluminium will heavily increase with electrification of vehicles. However, with today's recycling, contamination of aluminium alloys results in significant losses where wrought aluminium products are downcycled to cast aluminium with lower value and performance. This paper review the state of the art of aluminium recycling and investigate the current knowledge on the recyclability of current important aluminium alloys and their alloying elements. Future implementations and research are explored to find possible road maps for a sustainable circular economy of aluminium products. The findings indicate that closed-loop recycling trough better developed sorting and separation processes are one of the primary improvement directions. Also, improve utilization of the alloys and their alloying elements in the making of new aluminium alloys.

Keywords. Aluminum, recycling, literature survey, circular economy, impurities

1. Introduction

Today the circular economy is one of the significant trends in reaching a sustainable society [1]. One way to reduce climate impact is through increased recycling. With increased material recovery, less virgin materials are needed to provide the same benefits for the society. Metals are considered to have excessive sustainability benefits, as they, in theory, can be infinitive recycled. However, only a minor share of the metals in the recycling loops is in pure form and they are more common in alloys, composites and other material mixes. Mixed materials complicate the reuse and recycling processes as there is a risk of impurities and undesirable materials entering the material supply chain. Aluminium alloys, especially wrought aluminium, will heavily increase with the electrification of vehicles [2] due to its strength, lightweight and corrosion resistance.

¹ Corresponding Author, Christina Windmark, Production and Materials Engineering, Lund University, Ole Römers Väg 1, 221 00 Lund, Sweden; E-mail: christina.windmark@iprod.lth.se

Aluminium is a common metal in the earth's crust but considerable energy savings can be achieved from using recycled material compered to primary material [3]. However, with today's recycling, contamination of aluminium alloys results in significant losses where wrought aluminium products are downcycled to cast aluminium with lower value and performance. A study of recycling of aluminium-based car components shows that about 20 % virgin materials need be added to produce new cast aluminium products [4] and that about 45 % losses occur when aluminium is recycled. Most of the losses can be traced to the re-melting of the material [4]. This is an economic issue and a sustainability problem as the demand for wrought aluminium will increase the need for virgin materials, whereas the demand for cast aluminium in the vehicle business will decrease. Today about 35 % of all aluminium used in production in Europe is from recycling and it is estimated to be about 50 % in 2050 [5], which further increase the need for sustainable and efficient recycling. The need for well-functioning and value saving recycling togherter with remanufacturing actions [6] are therefore of high importance.

The aim of this study is to review the current practice of aluminium recycling, targeting a gap analysis for implementing a circular supply chain. Future implementations and research are explored to find possible roadmaps for a sustainable circular economy of aluminium products. The starting point in this paper is the EU standard SS-EN 13920 for grouping and sorting recycled aluminium dependent on the source, content, use, manufacturing methods and treatments. Different alloying elements are targeted to understand their importance better and understand the impact of their use. Purification techniques and implemented methods on recycling and using aluminium alloys to meet material specifications of new products are explored to find weaknesses and strengths in today's recycling activities. Different alloying elements are targeted to understand their importance better and their use.

2. Research Methodology

This paper is a review paper with the aim of finding the best practices and technology used for recycling of aluminium alloys as of today and to explore future recycling systems. This literature review will target three main areas: *sorting and classifications, alloying elements and their effect on recycling and material structures* and, *separation and purification techniques for recycling*. Different search engines, such as Scopus and Google scholars have been used for large coverage, with extra emphasis on material and material technology journals. Most searches has been focusing on aluminium alloys, but more general searches on material recycling has also been conducted. For each of the three different main areas, different subsearches have been used, using different search words, see Figure 1 for more information. In addition we also used snowballing, i.e. pursing references.



Figure 1. Search areas and main words.

3. Literature review

Recycled material can be divided into scrap from primary production and secondary materials from used products. One prognosis for 2050 is that the demand for aluminium will increase with 25 % and that most of this increase could be covered by increased use of recycled materials [5]. At present EU is a net exporter of aluminium scrap material, where about 80 % are exported to countries in Asia [7]. In 2019 EU exported over 1 million ton of scarp aluminium, which correspond to about 1/3 of the estimated annual aluminium scrap [8]. At the same time the union import about 14 million ton of aluminium (2018), where the main imports come from Switzerland, Norway and Iceland [5]. In an international perspective, the European aluminium production has low CO₂ emissions of about 18 kg CO₂/kg aluminium. This can be compared with the global average of about 18 kg CO₂/kg aluminium [5]. With this background, there are several incentives to further develop the European recycling and re-melting activities.

Aluminium is used in a wide range of different products within industries such as transportation, buildings, packaging, electronics consumer durables and machinery and the demand for lightweight applications with high strengths are increasing. To meet the increased demand and different products the aluminium industry is developing in the area of new alloys, joining technology, additive manufacturing and process improvements [5]. The development of additional alloys, the increased use of aluminium products, and increased mixes of materials imply that the complexity in recycling will increase and may affect the recycling efficiency negatively. The complexity also makes it important to use standards to better categorise and sort different alloys.

3.1. Scrap sorting and classification

The primary stage of recycling and one of the key components to realising sustainable recycling, is sorting of scrap. Re-melting losses and accumulation of impurities such as copper, ferrous materials, and oxides limits the number of times a general aluminium product can be recycled [9]. The complexity of the different types of alloys and application areas increase the possibility for impurities. The possibility to separate and purify is also limited and when used highly energy intensive which further add the importance to properly sort incoming materials [10]. To further support sustainable recycling, classification based on source, alloy content, manufacturing methods and

treatment are used [11,12] in combination with the European standard SS-EN 13920. In the European standard SS-EN 13920 there are 16 subgroups:

Part 1: General requirements, sampling and tests

Part 2: Unalloyed aluminium scrap

Part 3: Wire and cable scrap

Part 4: Scrap consisting of one single wrought alloy

Part 5: Scrap consisting of two or more wrought alloys of the same series

Part 6: Scrap consisting of two or more wrought alloys

Part 7: Scrap consisting of castings

Part 8: Scrap consisting of non-ferrous materials from shredding processes destined to aluminium separation processes

Part 9: Scrap from aluminium separation processes of non-ferrous shredded materials

Part 10: Scrap consisting of used aluminium beverage cans

Part 11: Scrap consisting of aluminium-copper radiators

Part 12: Turnings consisting of one single alloy

Part 13: Mixed turnings consisting of two or more alloys

Part 14: Scrap from post-consumer aluminium packaging

Part 15: De-coated aluminium scrap from post-consumer aluminium packaging

Part 16: Scrap consisting of skimming, dross, spills and metallics

The SS-EN 13920 cannot directly be connected to the recycling standards developed by the Institute of Scrap Recycling Industries Inc (ISRI) [11], however even as the two standards are defining scrap differently they together define available scrap and their impurities. The ISRI standard has more subgroups than the SS-EN 13920 and can therefore control post-consumer scrap and secondary materials depending on origin and treatment. However, all recycled scrap materials are to be transformed into alloys matching the standards required by the manufacturing industry such as the 1000-7000 series and 100.x-700.x series.

3.2. Alloys and impurities

Wrought aluminium alloys are divided into families based on the main alloying element: the 2XXX series is alloyed with copper, the 3XXX series with manganese, the 4XXX series with silicon, the 5XXX series with magnesium, silicon and magnesium are in the 6XXX series, and the 7XXX series is alloyed with zinc. The 1XXX series is the commercially pure aluminium. The Aluminum Association (AA) of the United States adopts a similar nomenclature for the cast alloys from 1XX.X to 7XX.X series [13]. Other elements are added for specific melt treatments: grain refinement is based on the addition of titanium and boron, while the eutectic modification is performed by adding sodium, calcium or strontium in the Al-Si systems. Beryllium can be added in traces to Al-Mg alloys because it reduces oxidation during melting [14]. Other elements are also present in these alloys as impurities, like iron, phosphorous and lead.

3.2.1 Alloying elements

There are several alloying elements important for the material properties for product function and manufacturing processes. Copper is the main alloying element in the 2XXX series but is also present in the Al-Si, Al-Zn and Al-Mg systems. Copper provides

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solution strengthening and makes the alloy heat-treatable with the precipitation of the θ -Al₂Cu phase [15] but also increases the hot tearing susceptibility of the alloy [16]. The addition of copper to the Al-Si systems promotes age hardening but negatively affects castability and corrosion resistance [14].

Manganese improves the mechanical properties of aluminium by solid solution and partially by dispersion hardening [14] and provides good corrosion resistance in different environments [15]. The addition of manganese to Al-Si systems that contain iron favours the formation of α -AlFeSi compounds. In this way manganese minimises the formation of large and brittle β -AlFeSi compound, and the Mn:Fe ratio should be 0.5:1 [14].

Silicon improves the fluidity of the molten metal and can be used for complex and finely detailed cast and forging products. The binary alloys are employed in applications which do not have strength as primary requirement, and the eutectic composition is particularly suitable for thin-walled products [14]. Magnesium and/or copper and nickel are often added to the binary system to increase strength by solid solution and precipitation hardening.

Magnesium provides solid solution strengthening and heat treatability [15]. The β -Al₈Mg₅ phase precipitates above the solubility limit, and it may determine intergranular corrosion and sensitivity to stress corrosion cracking. The addition of magnesium to the Al-Si and Al-Cu systems induces significant age hardening with the precipitation of the β -Mg₂Si phase and the S-Al₂CuMg phase, respectively [14].

Zinc combined with magnesium provides increase of strength by precipitation hardening of the η -MgZn₂ [14]. The stress corrosion cracking susceptibility can be improved with the addition of copper, at expenses of weldability.

3.2.2 Trace elements and Impurities

Titanium is added for grain refinement through Al-Ti or Al-Ti-B master alloys [14]. The compounds TiB2 and Al3Ti promote nucleation of the primary α -Al and thus determines a decrease in the average grain size. Titanium reduces hot tearing up to a content level where its refining ability is still active [17].

Sodium, calcium and strontium are added in traces to Al-Si systems for the eutectic modification treatment, which changes the morphology of eutectic silicon from flake-like to fibrous [18].

Chromium and nickel are not the main alloying elements in aluminium systems, but they are often present. Chromium is added in limited quantities to Al-Mg based systems to hinder grain growth and recrystallization during temperature exposure. It promotes the conversion of the detrimental β -Al5FeSi to the harmless α -Al15(Fe,Cr)3Si2 phase [19]. Nickel is added to increase strength, particularly at elevated temperatures thanks to the formation of load-bearing phases [20]. It combines with iron in the T-Al9FeNi phase [21].

Phosphorous is commonly present as an impurity in commercial purity aluminium and in the alloying elements for foundry alloys, leading to levels up to 20 ppm. Phosphorous forms aluminium phosphide (AIP) particles that act as nucleanting agents of silicon crystals [21].

Most alloying elements are difficult to remove but iron is of particular interest due to its wide use in combination with low tolerance for it in some applications. Iron is intentionally added to some alloys to improve the finished material properties (e.g. reduce germination and corrosion) or to facilitate the manufacturing and material processing (e.g. it enhances surface quality of die-cast products as these are not stuck on the die) [22]. Unintentional addition of iron includes sources such as the tools used during material processing. The disadvantage with iron is reduced ductility and tensile strength, since it makes it easier to form points of fractures. Primary aluminium alloys with low iron contents have entirely different use areas than secondary alloys. The first ones have superior properties related to strength, workability, weldability and corrosion resistance in wrought aluminium, and the second ones are employed in high pressure die casting as the iron is needed to avoid die-soldering [23,24]. Thus, iron plays an important role and affects the ability to produce high-performance alloys for recycling. Iron is detrimental for mechanical properties through the precipitation of several secondary phases: θ -Al13Fe4, Al6Fe, α -Al12Fe3Si, Al3Fe, τ -Al8Fe2Si and β -Al5FeSi [24,25]. When iron removal is not an option, its effects can be either limited with dilution with pure and low alloyed aluminium or controlled by the solidification parameters to obtain a targeted microstructure.

Das [26] concluded that aluminium alloys display a wide range of specific elements. The 2xxx series materials show wide ranges of copper, and the 6xxx series materials show wide ranges of magnesium and silicon, and so forth. The main problem is that the boundaries for impurities like iron, nickel, and vanadium required increased levels. Das [26] suggested that secondary alloy and application requires matching and suggested the following:

- 3xxx Heat-exchanger tubing, chemical piping
- 4xxx Forged or cast engine parts
- 5xxx Tankage plate; housing components
- 6xxx Extruded structural components

The challenges thus lie not only in the ability to receive and recycle but also in handling specific scrap types and identifying more tolerant applications that may conflict with market needs. Dilution to control iron and copper with pure material or 1xxx, 2xxx, 3xxx alloys is not feasible as it leads to a cost increase of the diluting material.

3.3. Purification and recycling techniques

Methods to use secondary aluminium in new products target different parts of the recycling process. Some of the methods overlap and/or can be combined. Here, we categorize the methods into the following six clusters that occur after scarp collection:

- 1. Closed loop recycling.
- 2. Sorting and preparing solid waste streams before melting.
- 3. Melt purification
- 4. Add reagents to neutralise impurities
- 5. Dilution with high purity aluminium and alloying elements
- 6. Downgrade or change new material specifications by widen acceptable material specifications.

3.3.1 Closed loop recycling

A resource efficient way to sort scrap and plan recycling is to do a closed-loop recycling, where products of the same alloying type is recycled into product of the same alloy. By closing the material loops it is, theoretically, possible to produce material that have the same material specifications and mass balance as the original material. If achieved, the main focus then can be on reducing energy consumption and increase the metal recovery rate [27]. However, in practice, this is difficult to realize as it is not possible to recover

100% of the contained material, tools coatings and recycling process steps always add

some impurities, material mixes are often used and oxides form during recycling. Even minor impurities will require treatment for consecutive cycles, since impurities accumulate [28].

3.3.2 Sorting and preparing solid waste streams

Sorting alloys into groups with similar alloying elements minimises material downgrading and the need to dilute impurities using virgin materials. Disassembly of products is typically expensive and material shredding is therefore used which results in waste streams with many different materials. These can be separated using magnets, conductivity (eddy current), colour sorting by hand or spectrographic techniques, measuring density, x-ray and/or 3D-shape [27]. However, these methods are inadequate for separating some material mixes that are joint together. So et al. [29] investigated joining techniques used in cars and found that unliberated mechanical joints contributed with 69% of the total iron impurities in their sample, the impurities mainly originated from screws, bolts and rivets. The authors suggested manufactures should be restrictive with the use of mechanical joints and recyclers to shred material into smaller pieces and then use strong magnets.

Sorting in itself do not separate the elements that are present in the alloys. However, sorting can enable higher utilisation of the alloying elements. Paraskevas et al. (2019) studied possibilities to sort scrap into groups that are as close as possible to the secondary materials that should be produced [30]. This maximises material utilisation as it reduces the need for both virgin aluminium and alloying elements. The study was theoretical and only included the four main alloying elements (iron (Fe), cupper (Cu), (Silicon) Si, and manganese (Mn)). Many more alloying elements are used and the proposed sorting methods may have limitations that where not addressed.

Material should be de-coated before melted. For example, using heat to remove paper, titanium oxides, lacquer, etc. This step is particularly important for achieving high recycling rates and closed-loop-recycling, as the coating will otherwise accumulate over multiple product cycles [30]. De-coating also improve accuracy for some sorting techniques and should then be done prior to these sorting processes, such as spectrographic techniques. When combined with chemical etching, de-coating enables separating the different alloy families (2XXX, 3XXX etc.) presented in 2.2 [31].

3.3.3 Melt purification

In general, it is difficult to remove impurities from molten aluminium. This is a result of aluminium's lower melting point, higher reactivity and higher oxygen affinity compared to its alloying elements. The only major exceptions to this among the common alloying elements are zinc (Zn) and magnesium (Mg) [32]. Magnesium (and calcium (Cr) and beryllium (Be)) can be removed by oxidisation and/or using fluxes that form stable chlorides and fluorides that can be removed. Zinc, cadmium (Cd) and mercury (Hg) can be removed by evaporation. According to Rao (2006), lithium (Li) can be removed by distillation at a fairly low cost [33].

Tin and lead have a lower melting temperature than aluminium and can therefore be removed by selective melting, i.e. keeping the metal at a temperature that is between the melting point of the contaminant and aluminium. Other elements remain in the molten aluminium and are therefore difficult to remove and thus it is difficult to reach a very high purity at a low cost.

3.3.3.1 Methods for removing elements from molten aluminium

There are several methods that are available, yet with some limitation issues, to remove impurities from molten aluminium including: gravity separation, filtration, electromagnetic separation, centrifugal separation (direct or from sludge), electrolysis, fractional solidification (or crystallisation) and fluxing refining [22].

Keeping the temperature of the molten aluminium at an elevated level (600-650°C) allows the formation of iron-rich intermetallics that can be removed as the iron rich phase fall to the bottom (gravity separation), are larger (filtration), heavier (centrifugal separation) or have lower conductivity and thus respond differently to electromagnetic forces (electromagnetic separation). Adding manganese (Mn), Cr, and Sr can increase sludge formation, thus enabling a lower iron content to be reached. These elements, and silicon, are removed as they are all part of the iron rich phase.

It is possible to combine the methods, e.g. gravity followed by filtration achieved an iron concentration of 0.27 wt.% [22]. Zhang et al. [22] finds that gravity separation, filtration and centrifugation are difficult to use for large scale volumes as they operate in batch (gravity), need cleaning (filters and centrifuges) and/or it is difficult to control temperature (centrifugal separation). The authors also conclude that electromagnetic separation has high potential to for commercial large-scale continuous processes (assuming that 0.4 wt.% Fe is acceptable). Electrolysis can be suitable if there is demand for very high purity, but the costs make it unsuitable for most applications.

3.3.3.2 Slag and dross recovery

In the dross valuable metals are accumulated among others are oxides from rare earth metals (REO) [34]. REOs content is low but is likely to increase in the future due to increased use of these elements in electronics and energy efficient electric conversion technologies. Thus, dross recovery is an important future activity. Zuo et al. [35] developed a multistage electrometallurgical method that was able to recover up to 97% of the valuable metals in the dross. The method consists of three steps: hydrolytic denitrification, calcination phase inversion and finally molten salt electrolysis. According to the authors, this method has the potential to be scalable to a low cost. Furthermore, it recovers more elements than currently used (hydrometallurgical) methods that mainly recover aluminium while as valuable alloying elements are wasted. Other methods under development include salt-free plasma technology, see Wibner et al. [34] for an overview.

3.3.4 Add reagents to neutralise impurities

The effects on material characteristics from impurities can, to some extent, be neutralised by adding reagents. This approach is manly used for impurities that are difficult to remove, such as iron. One of the main disadvantages with adding reagents is that the impurities are still present at about the same share and subsequent. Thus, recycling may accumulate their content to the extent that it compromises material specifications. As mentioned in 2.3.3.1, manganese is one of the most common reagents used to compensate for iron but many other are also suitable including cobalt (Co), chromium (Cr), beryllium (Be), strontium (Sr) and to some extent molybdenum (Mo), Nickel (Ni), and rare earths (RE) [22].

3.3.5 Dilution with high purity aluminium and alloying elements

This method compensates for quality losses taking place when, for example, different alloys are mixed during the recycling process. Both virgin and high-quality recycled

aluminium can be used. In the past, there has been a high demand for cast aluminium that require minor dilution when mixes of cast and wrought fractions are recycled. However, the biggest demand for cast alloys comes from internal combustion engine (ICE) vehicles (e.g. engine blocks) and a surplus of scarp cast alloys is therefore projected in the future as demand for ICE-vehicles declines [36]. This can drastically increase demand for virgin aluminium, unless more is functionally recycled and/or "upgraded".

3.3.6 Change new material specifications

In some cases, it is possible to change material specifications so that they can accommodate a larger share of impurities. Modaresi et al. [36] explored possible sink alloys within the automotive sector and found a limited number of suitable alloys. A handful of cast alloys were seen as most versatile, e.g. 301, 319 and A380. However, these alloys have a high content of alloying elements and may therefore come at the cost of increased demand for those elements. One additional factor identified by the authors is institutional barriers that restricts the use of recycled material. One example is aluminium wheels that are presently not recycled into new wheels due to safety standards.

4. Discussions and Future outlook

Scrap materials should not only be analyses based on its alloying content but also on the future performance requirements and costs to enable circular economy [37]. Looking at aluminium and aluminium alloys there are a large diversity related to area of use, and the alloying elements to support this use and the related manufacturing processes. To support the recycling of these products there are several main obstacles to consider. Based on the above it can be summarized to the following areas:

- Better techniques for shredding and sorting
- Closed-loop recycling of both primary and secondary materials to preserve material characteristics and higher material efficiency
- Better recovery of wrought aluminium and upgrading of scrap which can be obtained through:
 - A better alignment between existing scrap standards and the material specifications from manufacturers
 - Acceptance for more general specification when minimum quality aspects are reached design for recycling
 - Develop better purification techniques for removal of alloying elements

In addition to low collection rate, there are three major losses to consider in aluminium recycling: material losses, quality losses and dilution losses. These losses are a result of oxides and dross formation, quality losses due to downgraded material and dilution by adding new virgin material to the mix. For closed-loop recycling the latter two does not occur [38].

Das [26] studied the challenges in the design of a recycling-friendly alloy. Among the key challenges to be met in creating this ideal recycling-friendly alloy, one is to have the volume of a certain scrap. If there is not enough scrap in one category, it is hard to perform close-loop recycling or high value recycling of that particular alloys without adding new material. Another is to decrease the contamination by better pre-sorting, shredding and automated and optimized separation technology, which is in line with above findings. As a complement to today's separation and sorting techniques, X-ray Fluorescence (XRF), Laser-Induced Breakdown Spectroscopy (LIBS), and Prompt gamma neutron activation analysis (PGNAA) can be used, improving the accuracy of the process [27] and facilitate sustainable recycling.

Today the majority of aluminium recycling is conducted in open loops, which put pressure on developing the separation and purification techniques for the recycling to be sustainable. Nonetheless, recovery and separation of aluminium alloving elements is hard and energy demanding. To handle alloys with for example complex or high alloying content it is also crucial that the by-products can be utilized [26]. As an alternative Paraskevas et al. [38] shows in their study that there are considerable environmental gains to reduce quality losses and dilution losses by proper selection and utilization of scrap. When there is a need to add material to reach desired material mix the scrap utilization can increase and the need for primarily material decrease by using the composition of available scrap to gain the correct material characteristics. In this context, it should be mention that the economic incitement for more effective sorting is often not primarily based on the raw material price, but other driving forces such as higher material concentration and ese of separation can have a larger implication [39]. However, when recycling aluminium alloys the objectives is to produce an alloy fitting to the application and not pure aluminium. The findings above related to higher utilisation of the alloying elements trough sorting [30] indicate that the product design have an important role in managing an effective material recycling. This would also minimize contamination during different recycling processes such as shredding and melting.

A future perspective is to target control instead of cleaning: high shearing technologies enable the distribution of oxides finely and thus have more efficient nucleation of hydrogen in the final product [40,41]. A high-shear dispersive mixer is the base of a melt conditioned direct chill (MC-DC) developed at Brunel University London. This technology applied to the molten alloy before solidification produces a fine as-cast microstructure controlling irregular growth of intermetallic phases [42]. Chang et al. (2020) [43] applied this method to recycle AA6111 from different feedstocks and concluded that the mechanical properties obtained were promising without the need for chemical grain refiners. Al-Helal et al. [44] reported less segregation of iron-rich intermetallic in twin-roll cast AA6111. The uniform distribution of intermetallic particles obtained with high shear can increase the tolerance for impurities in recycled alloys. Lazaro-Nebreda et al. [45] recently demonstrated that the high shear melting conditioning could remove hydrogen and entrapped oxide films effectively, improving the quality of the melt.

5. Conclusions

The major shift in the automotive field to transform from combustion engine drive trains to electrification of vehicles makes it crucial to: 1) find a different application to absorb the end-of-life cast aluminium, and 2) limit further downgrading. In this paper the main obstacles for reaching circular economy for aluminium alloys have been summarized and possible future developments been investigated. The major drivers for recycling of aluminium are to reduce the use of energy, costs, environmental footprint and to increase the utilization of available resources. The technology for separation and purification needs further development to handle the material mixes of today. However, closed-loop

recycling with better sorting and more comprehensive standards, both for aluminium alloys and for aluminium scrap are pinpointed to be one of the important steps for sustainable recycling. Also, the possibility to utilize scrap as alloying incidents for producing aluminium alloys can be a promising way forward. One area not touched in this paper is the effect on recycling from low carbon primary aluminium production. Possibly the importance of the alloying elements then can be the driver for separation and/or reduced impact from mining in general (use of water, mine tailings etc.). Then, however, the technology needs to be developed for recovery of these elements.

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