10.1 Closed and Open Loop Recycling of Aluminium: A Life Cycle Assessment perspective

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Abstract

Compared to other base metals, the refining options for aluminium during the final metallurgical recycling stage are very limited. These limited melt purification options, along with the relatively large number of alloys and the accumulation of alloying and foreign elements during the different life cycle stages, force the aluminium industry to operate in a cascade recycling chain. A Life Cycle Assessment (LCA) based, resource oriented approach, is presented in order to: i) assess the environmental impact calculation during aluminium recycling, ii) examine the scrap recycling loops and iii) express and quantify dilution and quality losses during recycling. Finally, this paper discusses, from an environmental point of view, strategies and opportunities for improved recycling as well as opportunities for more sustainable scrap management. Case studies focusing on major post-consumer scrap streams are examined based on their environmental performance in two different recycling scenarios.

Keywords:
Life Cycle Assessment, Cascade, Aluminium Recycling, Resource Efficiency

1 INTRODUCTION

Compared to other material categories, metals have the highest potential for systematic recycling, due to i) their high economic value, ii) their large volumes enabling economies of scale and iii) their distinctive feature of excellent recyclability. Nevertheless, many challenges related to the accumulation and mixing of alloying elements due to inefficient alloy separation and accumulation of impurity elements (also known as tramp elements) during the different life cycle stages, along with the removal limitations of certain elements during smelting processes, are still not properly addressed.

Except for aluminium foil, aluminium is not used in its elemental form, but rather as an alloy. More than 450 alloy designations/compositions have been registered by the Aluminium Association Inc. [1]. Typical alloying elements depending on the alloy series are: Si (Silicon), Cu (Copper), Zn (Zinc), Mg (Magnesium) and Mn (Manganese), with Fe (Iron) occurring mainly as an impurity element. Two major categories are defined with respect to the concentration of alloying elements: i) high purity wrought alloys (with alloy content up to 10%) and ii) cast alloys with much higher tolerance limits for alloying elements (with alloy content up to 20 wt.%), especially for Si. The relatively broad variety of alloys, combined with the often inefficient pre-melt alloy separation, leads to higher entropy scrap streams (a mixture of several alloys) that are difficult to handle.

Nowadays, this challenge is successfully addressed either by dilution of the difficult to handle impurities with high purity metal inputs, or by cascade recycling to alloys with lower purity requirements. Since there is limited scrap availability and at the same time a high demand for highly alloyed products (cast and die cast alloys), both strategies effectively address this industrial problem. Nevertheless, one can wonder whether these strategies will remain sustainable solutions in the future.

2 CHALENGES IN ALUMINUM RECYCLING

2.1 Refining options and limitations

Recent studies [2,3], based on chemical thermodynamic analysis simulating smelting processes, indicate which elements can be removed and in how far impurity can be controlled during recycling of various base metals, such as, aluminium, steel, copper and magnesium. Nakajima et al, [2] examined the distribution of 45 elements (most of them occur as tramp elements) in the gas/metal and slag/metal phase for simulated aluminium remelting. In most cases melt purification can be performed either through the oxidation mechanism in the slag phase or by evaporation in the gas phase. The authors concluded that, amongst the examined elements, only Mg, Ca and Be can be removed in the slag phase, and Zn, Cd and Hg in the gas phase under varying oxygen partial pressure and temperature conditions. Regarding the contamination by the typical alloying elements, only Mg (in the slag phase) and Zn (in the gas phase) can be removed to an appreciable extend. The residual 39 elements remain in the metal phase. In consequence the purification of the melt from the elements that remain in the metal phase during the final recycling step processes is either very difficult or essentially impossible to achieve. Moreover, while a post electrolysis process can recover most of the impurity elements that remain in the metal phase during smelting of copper, this is not the case for aluminium. Compared to the primary production of aluminium, consuming, approximately

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14 kWh/kg, the three layer electrolytic process for aluminium refining is more energy intensive with a consumption of 17-18 kWh/kg [4].

Flux treatment is the most common and widely used melt liquid metal treatment in industry since it: i) reduces oxidation, ii) removes hydrogen and nitrogen gases, and iii) removes Ca, Sr, Na, Mg and Li inclusions from the melt [4]. Moreover, a flotation/de-gassing treatment of the melt, purging gases containing chemical reactive components such as chlorine gas, can also be an effective solution in removing Na, Ca, Li and Mg apart from hydrogen [5]. Zinc, a major alloying element in the 7XXX alloy series, can be recovered using distillation technologies [4]. Finally, other technologically advanced melt/chemical separation technologies, like fractional crystallization and unidirectional solidification, are still in a research or early development stage [5]. Thus, from technical and economic point of view, these technologies are still questionable and not proven to be viable for scrap purification at large scales.

2.2 Scrap Sorting

New scrap from production can be separated into two categories according to its source: i) semi-manufacturing and ii) manufacturing scrap. The forming scrap from the semi-manufacturing sector (e.g. extrusion and rolling scrap) concerns relative large and homogeneous flows where the benefit of knowing the chemical composition is partially utilized by recycling internally by integrated cast houses. These scrap streams are usually not available in the scrap market. Thus, more opportunities for more sustainable scrap management and added scrap value by sorting can be found in the product manufacturing scrap and in urban mining where the composition uncertainty due to alloy mixing is at higher levels. Inefficient pre-melt scrap sorting in product systems containing both fractions (like automobiles) will result in the wrought alloy fraction to be lost in the cast fraction. Table 1 presents the Si content of blended wrought and cast alloys for various mixing ratios. By mixing wrought/cast alloys in increasing ratios, the concentration of Si in the scrap stream is increased and thus lowering the scrap quality in terms of recyclability. Increased concentrations of a contaminant results in limited options for cascade or higher dilution needed to up-cycle to higher purities leading to underutilization of the scrap. Alloying elements, such as Si, are essential elements; but the when different alloys are mixed in an uncontrolled manner, they can became contaminants.

Table 1: Average Si content for various blended wrought cast fractions [5].

Wrought/Cast ratio	Average % Si
1/5	8.0
1/1	5.0
2/1	3.5
5/1	2.0
10/1	1.3

The recovery of embedded financial value of the scrap stream composition can be positively affected by a recycling strategy based on separate recycling of different alloy categories/series. Taken into account also the economic benefits from this separation, regional standards are being established in order to control the scrap quality. In the European Community, the DIN EN 13920 standard [6] identifies 15 different categories of aluminium scrap. Similar scrap categorisation systems, but focusing more on scrap from different product systems, can be found also in other countries [7]. Thus, any scrap dealer or trader supplying sorted scrap according to these standards can set a higher price and therefore obtain a commercial advantage. Moreover, the impact of scrap usage is inversely proportional to the production cost and by increasing scrap utilization the overall production cost can be reduced [8].

2.3 Recycling Circuits and Options

To resolve the challenges linked to poorly managed scrap streams, the Aluminium industry makes use of a cascade recycling chain. In Figure 1a a simplified representation of an aluminium cascade chain is depicted. A graphical representation of the pathways for industrial scrap (yield loss of the production), and for post-consumer scrap according to their recycling options is shown in Figure 1b.

Unalloyed/low alloyed Al (1XXX series), coming from primary Al, can be considered at the top of the chain as high purity (in terms of Al content) products. Wrought alloys are in the middle of the chain, while alloys with low purity requirements alloys act as a sink in the cascade recycling stage. Since wrought alloys have strict and very low tolerance limits for alloying elements, their production is heavily dependent on primary Al consumption. Mixed alloy scrap streams are difficult to be absorbed into a wrought product due to mixing and excess concentrations of alloying elements. For that reason downgraded or mixed alloy scrap streams are mainly diverted to cast alloys. A common practice nowadays is to down-cycle mixed wrought alloys to cast quality. For primary Al there is no restriction of utilizing it to produce any alloy by adding alloying elements, and since there is limited scrap availability, primary Al is used as high purity metal input to balance the cascade chain. Figure 1a,b represents the recycling options and the recycling loops of the scrap streams according to the quality of the input/output (I/O) metals that are investigated from an environmental perspective. These recycling options are:

Preserving quality **:** Meaning that the quality of the scrap streams is effectively controlled and recycling is performed in a compositionally closed recycling loop, without any significant change in the inherent properties (alloy content) between the input and output materials. The single alloy strategy (illustrated by the green line in Figures 1a,b, represents the completely closed compositional recycling loop, effectively avoiding the need for dilution and quality degradation. The environmental impact per kg of produced alloy is minimal and can be mainly attributed to the energy requirements of the recycling process itself.

Downcycling : Accumulation of impurities and alloying elements to alloy products with lower purity requirements, when higher purity secondary resources are used to produce lower purity products (e.g. transition from mixed wrought alloys to cast quality).

Upcycling: When impurities are diluted with high purity metal inputs. Taking into account the limited scrap availability and high demand for cast alloys, the dilution agent is primary Al, especially for wrought aluminium production.

From economic point of view the high purity metal in the case of dilution, as well as the reduced product value in the case of

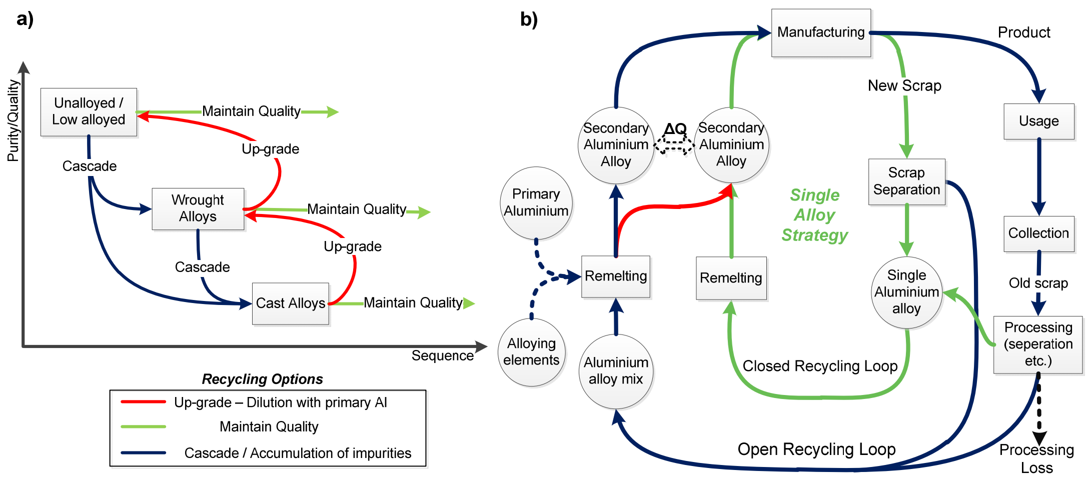

Figure 1: a) Al Cascade recycling chain and b) Recycling options and corresponding pathways.

down-cycling form incentives towards more efficient quality preserving solutions.

2.4 Resulting Challenges

In recent years increased concern has risen from researchers regarding the sustainability of the currently used industrial recycling strategies. A recent study [9] indicates that the introduction of electric vehicles will result in a decrease of the demand for cast alloys, generating 6.1 Mt of scrap in 2030 which will not be recycled due to the high concentration of alloying elements. The same research group forecasts a surplus of 12.4 Mt of scrap with high alloy content in 2050 that will not be able to be recycled. Modaresi and Muller [10] developed a dynamic material flow model at a global scale for the automotive system. They concluded that the continuation of the above mentioned strategies will result in a non-recyclable scrap surplus by around 2018 with an uncertainty margin of about 5 years.

The mixture and accumulation of alloying elements in the final scrap streams, especially those for which the removal from the melt is problematic, indicates that besides the amount of Al scrap also the composition must be taken into account in the environmental impact assessment at the material recycling phase.

3 METHODOLOGY

A major bottleneck in current LCA methods is that quality degradation during recycling cannot be properly addressed [11]. Most studies account the scrap flows and the produced secondary metals as equivalent in terms of quality, ignoring quality degradation. Recent studies highlight the role of quality and dilution losses during metal recycling, focusing on streams of ferrous materials [12] and aluminium [11]. In this paper the term quality is referring to purity or Al content of the stream and not to the physical properties.

For this purpose and in order to assess the environmental impact during aluminium recycling, an LCA based methodology has been developed. This approach takes into explicit account the interconnection between the quality of the resource inputs and the specifications of the alloy product focusing on the contamination by alloying elements during Al recycling. The environmental impact is used as a metric to express and quantify quality and dilution losses during metal recycling. The presented methodology can be used to evaluate the efficient use of resources during the secondary metal production. Figure 2 presents the system boundaries and functional unit of the LCA based analysis. Since the secondary production process aims at a specific aluminium alloy as output, the functional unit is set at 1 kg of the target aluminium alloy. In order to accurately quantify and assess the environmental impact of the production of the target alloy, the composition of the input scrap streams as well as the desired target alloy specifications (compositional tolerance limits according to the industrial standards [1]) need to be taken into account. Primary Al needed for dilution of the hard to refine alloying elements of the scrap mixture and required additions of all the typical alloying elements in order to adjust the alloy composition after dilution were included.

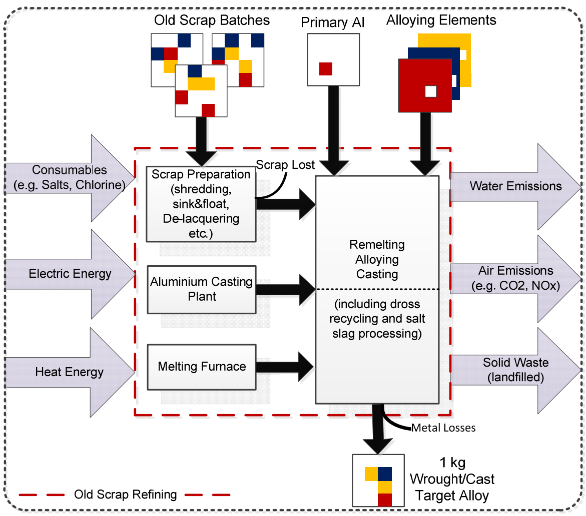

Figure 2: System boundary of the Al recycling process including melt refining and the functional unit.

The analysis is focusing on the four major alloying elements (Si, Fe, Cu and Mn) for Al that remains in the metal phase during remelting [2]. For these elements, the removal from the melt is very difficult or technically impossible, as explained in Section 2.1 and dilution is required to decrease their concentration in the melt. Mg was excluded; since it can be easily extracted from the melt (in the slag phase) at some appreciable extend by the widely industrially used salt fluxing and flotation/de-gassing refining technologies [5,6]. The environmental impact of these treatments along with the dross and salt slag recycling were included. The ReCiPe LCIA method (Endpoint Europe H/A) was used to calculate the environmental impact. The Eco-Invent v2.2 LCI datasets [13] and the latest environmental report from the European Aluminium Association (EAA) [14] were used to model the processes. The environmental impact is expressed in "ecopoints" Pts, (1000 mPts=1 Pts), where one point can be interpreted as one thousandth of the annual environmental load (damage) of one average European inhabitant.

Zinc is the major alloying element only for the 7XXX alloy series, and for the other series occurs in low concentrations. The influence of Zinc was considered to be negligible since: i) can be partially lost by evaporation in the gas phase, and ii) the 7XXX alloy series was not included in the case studies. From the EEA recycling model [14], it is estimated that 1108 kg of Al scrap enters the scrap preparation phase (consisting of unit processes like shredding, shink and float, de-lacquering) accompanied with approximately 211 kg of foreign materials. Afterwards, 1055kg of scrap exits the scrap preparation and enters the melting model for producing 1 tonne of ingot. Material losses which include the scrap preparation losses and the melting losses are also included.

4 CASE SELECTION

The investigated scrap streams are examined based on their expected mean compositional values from the literature and are considered to be discarded from: i) castings used in engines and transmissions from vehicles [8], ii) wrought products from automobiles with a rough separation between cast and wrought fraction [15], iii) wire and cable scrap [6], iv) used beverage cans (UBC) [6], v) buildings [9], vi) consumer durables [9], vii) from the major alloy series 6XXX excluding the two main alloys of this series AA6061 and AA6063 [9] and viii) a single alloy AA6061 [1]. To handle scrap composition uncertainties, the compositional window of the target alloys (maximum and minimum tolerance limits) for each element was narrowed by 5%. Al 99.7 (1070 AA similar alloy) is considered equivalent to primary Al, according to the chemical composition specifications of the London Metal Exchange for high grade aluminium contracts [16].

As target alloys in the case studies the following materials were used: AA3104, one of the largest volume alloys in the industry since it is used in the can body of beverage cans among other applications, and ii) 380.0, a typical die cast alloy used in automobile castings with high composition limits in alloy content.

A graphical representation of the mean/expected composition values of the scrap streams and of primary Al used as diluting agent, along with the target alloys' maximum composition tolerance limits, can be found in Figure 3. Each scrap batch was used as input, together with primary Al and alloy elements (assuming sufficient quantities), to examine: i) the maximum scrap utilisation of each scrap stream, ii) the normalised overall environmental impact per normalised kg and iii) the quality and dilution losses. The maximum scrap utilization rates of each scrap stream for the production of the selected target alloys is presented in Figure 3.

5 CONCLUSIONS

The environmental impact contributions per kg of the desired target alloys, representing the environmental optimum scenario of maximum scrap utilization and minimum material down-cycling for the desired alloy production, are shown in Figure 4. Based on the results the following major conclusions can be drawn:

- The engine and transmissions batch (cast alloys), having a high alloy content, cannot be utilised in large quantities to produce the target alloy AA3104. The low scrap utilization value (7.8%) in most cases is the reason why cast alloys cannot be recycled to anything else than casting quality. In contrast, when the target alloy is the AA380.0, specifically used for vehicles casting, the impact lowers significantly since no dilution is needed except for a small addition of alloy elements. The scrap usage in the latter case reaches 99.7%.

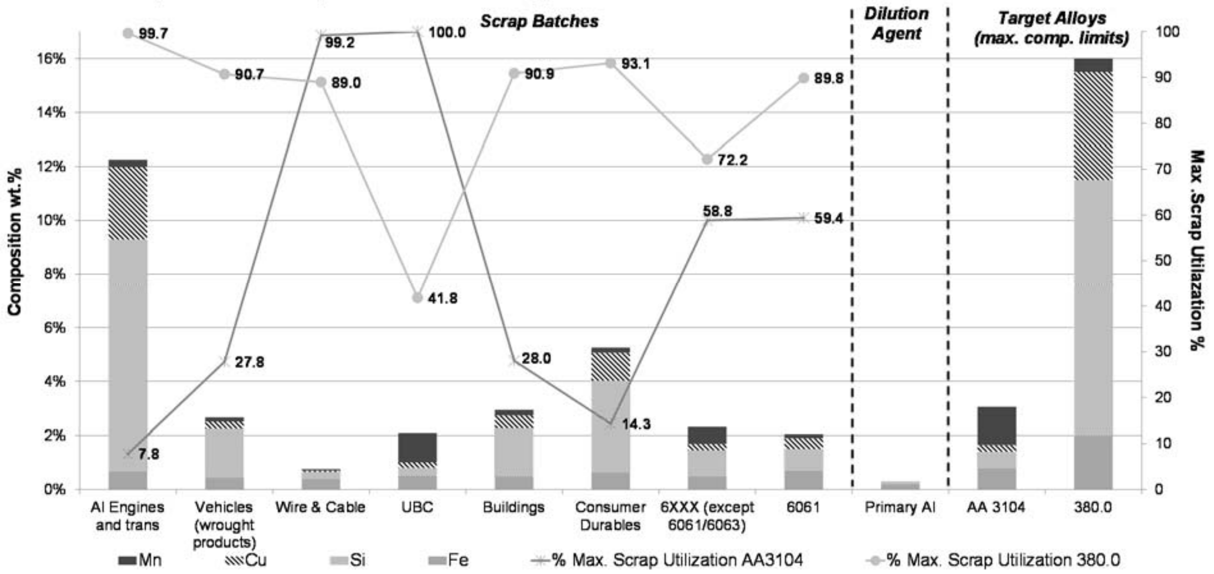

Figure 3: Composition of the examined scrap batches, primary Al along with the maximum scrap utilization for each target alloy.

- Preserving the quality of the scrap streams (UBC scrap to AA3104 or engine scrap to auto cast alloys) by utilizing it in compositionally closed recycling loops, can offer significant environmental benefits. UBC scrap can be fully utilised in the AA3104 alloy (used for the can tabs) without any addition of alloying elements and primary Al reaching the optimum 100% scrap usage. This strategy is already followed and represents an ideal example of an appropriate closed loop recycling based on separation of the scrap streams according to product applications. But this strategy is not always followed and can be expanded to other product systems, e.g. Al engine scrap to an alloy used for automotive castings.
- A ‘rough’ separation [15] between cast and wrought scrap in the End-Of-Life (EoL) treatment of vehicles can be environmentally beneficial. In particular, during the EoL treatment of vehicles, the engines are removed from the car in order to be depolluted. Thus, the biggest cast fraction in automobiles can easily be sorted separately and used in the auto cast production with higher scrap utilisation than wrought alloys, substituting the alloy elements addition with the alloy content. Hatayama et al. [9] illustrated that scrap sorting of ELV can significantly lower the future generation of un-recycled scrap and reduces the primary Al requirements by 15-25%. The separated wrought fraction can be used more efficiently for wrought alloys production, increasing the old scrap usage and decreasing the primary Al consumption.
- The scrap streams from: i) buildings and ii) wrought products from vehicles show a similar impact performance since they are characterised by only limited compositional differences. By selectively collecting them and utilizing them for wrought alloy production, higher substitution values for primary Al can be achieved.
- Depending on the desired target alloy, a suitable scrap input can be identified from environmental perspective. Moreover, it is possible to also identify how much scrap sorting is needed depending on the targeted alloy output. For the production of the AA3104 alloy, for example, the 6XXX and the 6061 alloys have similar maximum scrap utilisation. Thus, the sorting strategy in that case can target the 6XXX alloy family and additional sorting may not be required within this alloy series.

Quality and Dilution losses during recycling

A graphical representation of the overall environmental impact, combined with a breakdown analysis of the impact contributions, is shown in Figure 4. The overall environmental impact can be separated into four different contribution areas: i) the impact of the recycling process itself (mainly due to the energy consumption) and excluding the metal inputs ii) the scrap usage impact consisting of the scrap preparation and transportation to the plant, iii) the impact of the primary Al addition needed to dilute the contaminants, and iii) the impact of the alloying elements addition. The impact of the latter areas represents the dilution and quality losses respectively. Dilution of the impurities can be made also with high purity scrap inputs, but scrap blending scenarios will be examined in future work and was considered out of the scope in this analysis in order to examine each scrap stream separately. Dilution loss of the scrap mixture is expressed by the environmental impact originating from the primary Al addition and quality loss is reflected by the alloy elements addition. A graphical representation of these losses can be found in Figure 4, for the two representatives and at the same time contradictory case studies (wrought versus cast alloy).

For the production of the AA3104 alloy, most of the scrap batches need to be diluted and quality loss does not contribute significantly to the overall impact per kg. On the other hand, targeting the lower purity requirements of the 380.0 alloy, quality losses are visible highlighting the significance of material down-cycling during recycling.

Targeting the 380.0 alloy as output, none of the scrap batches, except for the UBC and the 6XXX, need dilution. The difference in scrap usage (from 99.7% for the engine and transmission scraps to the 89.0% scrap usage of the wire and cable scraps) is due to the alloying elements addition that can

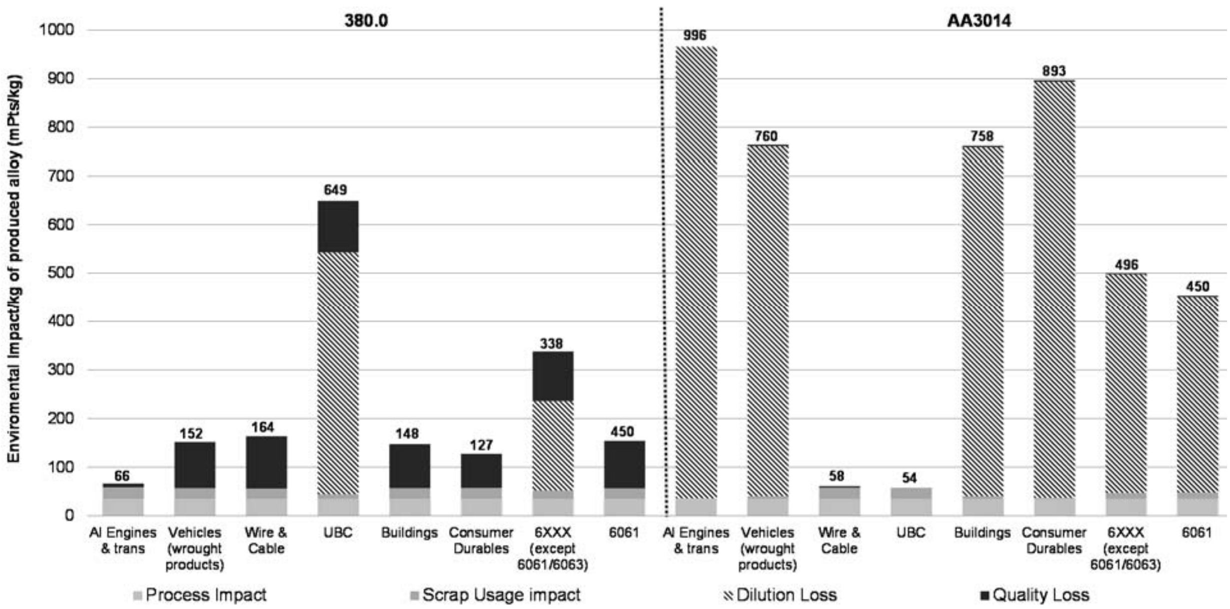

Figure 4: Overall environmental impact/kg (mPts/kg) along with the impact contributions for the production of the target alloys utilizing each scrap batch, primary Al and alloying elements.

significantly influence the overall environmental impact per kg of the produced alloy. Due to the higher quality losses, the wire and cable scrap batch shows a nearly 2.5 times higher environmental impact per kg (152 mPt/kg), compared to the engine and transmission scrap batch (66 mPt/kg). This example highlights the importance of minimising material down-cycling at production/product level.

Utilizing high purity and well defined scrap streams in wrought alloy production, the overall environmental impact can be significantly reduced by decoupling wrought production from primary Al at higher rates. Improving the sorting of post-consumer scrap, the wrought fraction can be increased, resulting in more efficient substitution of primary Al in the wrought alloy production. Moreover, since quality loss is an important impact contributor; the effort should be to substitute alloying elements addition with the scrap alloy content.

6 OUTLOOK

Since the removal of alloying element during remelting is very difficult or problematic for most of the elements, it is important to more efficiently control their concentration in the scrap streams before re-melting. Comparison of the environmental impact to process scraps metal back into different target alloys, is crucial to develop a sustainable scrap management strategy and identify compositional tighter recycling loops. In this respect the single alloy strategy is the best sorting approach since quality loss and the need for dilution are avoided. Utilizing high purity scrap streams for the production of wrought alloys, higher primary Al substitution rates can be achieved. Furthermore, lower purity scrap streams have a higher potential to substitute alloying element addition as require in cascade recycling.

Moving from open to compositionally ‘tighter’ recycling loops, finally resulting in single alloy recycling loops, will minimize the need for primary Al required for dilution and the need for alloying elements addition, resulting in large environmental and economic improvements at production and ultimately product level. Finally, a more different approach focusing on solid state / meltless recycling of aluminium scrap can provide significant environmental benefits, mainly by avoiding metal losses during remelting [17].

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