NTNU – Trondheim
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Life cycle assessment of secondary aluminium refining

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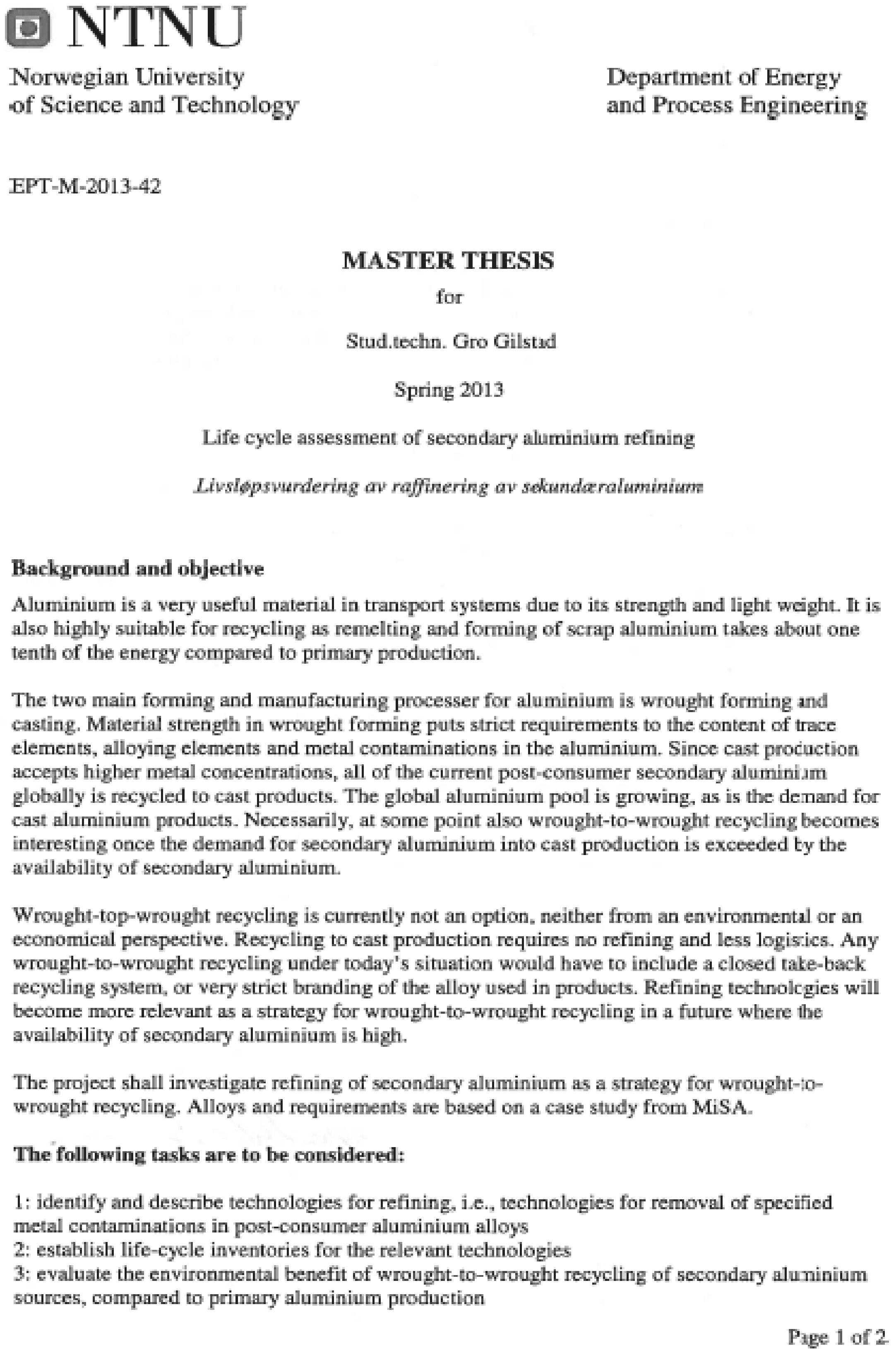
Problem description



NTNU

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and Process Engineering

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MASTER THESIS

for

Stud.techn. Gro Gilstad

Spring 2013

Life cycle assessment of secondary aluminium refining

Livsløpsvurdering av raffinering av sekundæraluminium

Background and objective

Aluminium is a very useful material in transport systems due to its strength and light weight. It is also highly suitable for recycling as remelting and forming of scrap aluminium takes about one tenth of the energy compared to primary production.

The two main forming and manufacturing processer for aluminium is wrought forming and casting. Material strength in wrought forming puts strict requirements to the content of trace elements, alloying elements and metal contaminations in the aluminium. Since cast production accepts higher metal concentrations, all of the current post-consumer secondary aluminium globally is recycled to cast products. The global aluminium pool is growing, as is the demand for cast aluminium products. Necessarily, at some point also wrought-to-wrought recycling becomes interesting once the demand for secondary aluminium into cast production is exceeded by the availability of secondary aluminium.

Wrought-top-wrought recycling is currently not an option, neither from an environmental or an economical perspective. Recycling to cast production requires no refining and less logistics. Any wrought-to-wrought recycling under today's situation would have to include a closed take-back recycling system, or very strict branding of the alloy used in products. Refining technologies will become more relevant as a strategy for wrought-to-wrought recycling in a future where the availability of secondary aluminium is high.

The project shall investigate refining of secondary aluminium as a strategy for wrought-to-wrought recycling. Alloys and requirements are based on a case study from MiSA.

The following tasks are to be considered:

1: identify and describe technologies for refining, i.e., technologies for removal of specified metal contaminations in post-consumer aluminium alloys
2: establish life-cycle inventories for the relevant technologies
3: evaluate the environmental benefit of wrought-to-wrought recycling of secondary aluminium sources, compared to primary aluminium production

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Within 14 days of receiving the written text on the master thesis, the candidate shall submit a research plan for his project to the department.

When the thesis is evaluated, emphasis is put on processing of the results, and that they are presented in tabular and/or graphic form in a clear manner, and that they are analyzed carefully.

The thesis should be formulated as a research report with summary both in English and Norwegian, conclusion, literature references, table of contents etc. During the preparation of the text, the candidate should make an effort to produce a well-structured and easily readable report. In order to ease the evaluation of the thesis, it is important that the cross-references are correct. In the making of the report, strong emphasis should be placed on both a thorough discussion of the results and an orderly presentation.

The candidate is requested to initiate and keep close contact with his/her academic supervisor(s) throughout the working period. The candidate must follow the rules and regulations of NTNU as well as passive directions given by the Department of Energy and Process Engineering.

Risk assessment of the candidate's work shall be carried out according to the department's procedures. The risk assessment must be documented and included as part of the final report. Events related to the candidate's work adversely affecting the health, safety or security, must be documented and included as part of the final report. If the documentation on risk assessment represents a large number of pages, the full version is to be submitted electronically to the supervisor and an excerpt is included in the report.

Pursuant to "Regulations concerning the supplementary provisions to the technology study program/Master of Science" at NTNU §20, the Department reserves the permission to utilize all the results and data for teaching and research purposes as well as in future publications.

The final report is to be submitted digitally in DAIM. An executive summary of the thesis including title, student's name, supervisor's name, year, department name, and NTNU's logo and name, shall be submitted to the department as a separate pdf file. Based on an agreement with the supervisor, the final report and other material and documents may be given to the supervisor in digital format.

☐ Work to be done in lab (Water power lab, Fluids engineering lab, Thermal engineering lab)
☐ Field work

Department of Energy and Process Engineering, 14. January 2013

Olav Bolland
Department Head

Edgar Hertwich
Academic Supervisor

Research Advisor: Johan Pettersen, MiSA as

Preface

This work was completed in the spring of 2013 to fulfil the Master thesis requirement of the Environmental and Energy program at NTNU.

Through the work with this project I have had to put in use, and combine, my theoretical background from various courses. I have especially used knowledge from courses containing chemistry, thermodynamics, basic math and the LCA methodology. At the same time I have learnt about a new field; metallurgy and the entire industry of recycling of aluminium. During this work I have seen LCA from a new perspective by experiencing the laborious work of searching for good inventory data that only exists within a limited range, and trying to close the gap between theoretical knowledge, future projections and real life practise. And it has indeed been both challenging and interesting!

I want to share my deepest gratitude towards Anne Kvithyld for showing sincere interest in my work with this thesis and for sharing her knowledge and network of brilliant co-workers. I believe I would not have gotten this far without her interest and enthusiasm.

I would also like to thank my supervisor Johanne Hammervold at MiSA for sharing her experience, listening to my worries and providing insightful feedback on my work.

I would also like to share my appreciation towards my entire family and friends, for their endless support and encouragement throughout the duration of my entire education. They have been there as moral support, as discussion partners and as babysitters whenever needed. Thank you! A very special mention to my partner in crime, John, you are the best. Your support is indescribable and this would have been impossible without you. I am forever grateful.

It is my sincere hope that this research will be useful for further use in the SupLight project and to anyone else who finds this topic interesting.

Trondheim June 28th 2013.

Gro Gilstad

«I would like to dedicate this work to my beautiful son, in the hope that through small steps of improvements we will be able to create a better world for future generations».

Abstract

The goal of this life cycle assessment is to investigate refining methods which makes it possible to utilise post-consumer aluminium scrap in production of aluminium products that have strict requirements to chemical purity. A further goal is to establish whether applying these methods are environmentally preferable compared to using primary aluminium to produce the same products. The investigation is based on a given scrap composition and given purity limits that must be achieved for the refined aluminium, related to six alloying elements. Based on the given requirements, a selection of refining processes which are able to refine the given scrap to meet the given limits are identified. Table A below gives the scrap composition and the desired range of removal to fit seven different alloy requirements. The scrap composition is given in weight%.

Table A: Scrap composition and desired level of removal.

	Si	**Mg**	**Fe**	**Cu**	**Mn**	**Zn**
Scrap composition	6.11	0.53	0.53	1.66	0.23	0.86
Desired removal	10 – 83 %	-	6 - 57 %	45 - 100 %	-	77 - 100 %

Different types of electrolysis and fractional crystallisation are identified as possible refining processes. Electrolysis can in theory be used to eliminate any tramp element in the scrap, as it extracts the pure aluminium from the melt using an electric current. Unfortunately this is an expensive, energy consuming process which requires use of chemicals. Fractional crystallisation does not require any chemicals and generally has low energy use, but this is a slow method which currently is not continuous. The ability of fractional crystallisation to remove the various elements depends on their solubility in aluminium in solid and liquid state. Various studies show that this method is promising for removal of Si, Fe and Cu, mediocre for Mg and Zn and poor for Mn. Based on this, six different production scenarios for secondary scrap, to meet the set of alloy requirements, were developed.

The life cycle impact assessment conducted for the different refining scenarios shows that energy use is very closely linked to the environmental burden associated with each of the production scenarios. From an environmental perspective low temperature, electrolysis and fractional crystallisation seem to be the best alternatives. Since the assessment is based on specific requirements set by the scrap composition and the given purity limits, an overall impression is that other possibilities to handle excess scrap should be investigated further. For example better sorting processes to separate tramp elements earlier in the production or development of alloys which are made with a motive for recycling. Such methods are likely to be more relevant when the use of aluminium has increased even further and more stable sources of scrap can be established.

Sammendrag

Målet med denne livsløpsanalysen er å undersøke raffineringsmetoder som gjør det mulig å benytte aluminiumskrap i produksjon av aluminiumsprodukter som har strenge krav til kjemisk renhet. Et videre mål er å vurdere, ut ifra et miljøperspektiv, om å benytte disse metodene er å foretrekke fremfor bruk av primær aluminium. Analysen er basert på en gitt skrapsammensetning og gitte renhetsgrenser som skal oppnås for det ferdig raffinerte metallet, relatert til seks legeringselementer. Basert på de gitte kravene er mulige raffineringsmetoder identifisert. Tabell A gir en oversikt over skrapsammensetningen og ønsket intervall for fjerning av legeringselementene. Skrapsammensetningen er gitt i prosent av total vekt.

Tabell A: Skrapsammensetning og ønsket intervall for fjerning av legeringselementene.

	Si	**Mg**	**Fe**	**Cu**	**Mn**	**Zn**
Skrapsammensetning	6,11	0,53	0,53	1,66	0,23	0,86
Ønsket fjerning	10 – 83 %	-	6 - 57 %	45 - 100 %	-	77 - 100 %

Tre-lags elektrolyse, lavtemperaturelektrolyse og delvis størkning er identifisert som mulige raffineringsmetoder for dette skrapet. Elektrolyse kan i teorien benyttes til å fjerne alle uønskede elementer i aluminiumskrap, siden denne prosessen går ut på å trekke ut det rene aluminiumet fra skrapet ved hjelp av elektrisk strøm. Problemet med denne metoden er at den er dyr, har et høyt energiforbruk og benytter ulike kjemikalier. Delvis størkning benytter ingen kjemikalier og har generelt et lavt energiforbruk, men dette er en langsom prosess som foreløpig ikke er kontinuerlig. Ikke alle uønskede elementer kan fjernes ved delvis størkning. Ulike studier viser at fjerning av Si, Fe og Cu kan være god, middelmådig for Mg og Zn og dårlig for Mn. Basert på dette er seks ulike produksjonsscenarier som møter renhetskravene utviklet.

Livsløpseffektvurderingen viser at energibruk er sterkt relatert til miljøpåvirkningen som assosieres med de ulike produksjonsscenariene. Fra et miljøperspektiv vurderes lavtemperaturelektrolyse og delvis størkning til å være de beste alternativene. Med tanke på at livsløpstolkningen er basert på en gitt skrapsammensetning og gitte grenser som skal oppnås for legeringselementene, er det opparbeidet et inntrykk av at andre muligheter for å håndtere overskuddsskrap også bør vurderes. For eksempel ved å forbedre sorteringsmetoder for å skille ut uønskede elementer tidligere i produksjonen eller ved å utvikle legeringer med tanke på resirkulering. Slike tilnærminger antas å bli mer relevante dersom bruk av aluminium fortsetter å øke og mer stabile kilder til skrap etableres.

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Terms and abbreviations

Alloying element	An element added to the aluminium to give desired properties to the metal.
Anode	Positive electrode in electrolysis, where reduction occurs (element receives electrons).
Binary solution	Solution composed of two elements, in this context aluminium with one other element.
Cascade recycling	Recycling to a lower-value product.
Cast alloy	Alloys formed by solidification of molten state, and often allow higher alloy and impurity content than wrought alloys.
Cathode	The electron supplying electrode in electrolysis, where oxidation occurs.
Closed-loop recycling	One type of products are collected and recycled back to the same products.
Continuous process	The supply of scrap and removal of pure aluminium is continuous.
Cu	Copper.
Dilution	Describing when primary aluminium is added to scrap melt to dilute it, to meet purity levels.
Downcycling	See cascade recycling.
Dross	Solid impurities either floating on top of the molten metal, or sometimes dispersed in the melt. Often includes oxides.
EAA	European Aluminium Association.
Electrolysis	A process where an electric current is used to force an otherwise non-spontaneous reaction.
Electrolyte	A substance which is soluble in water and produce ions which can conduct an electric current (Zumdahl, 2005). Used in electrochemistry to transport electrons between anode and cathode.
ELV	End of Life Vehicles (vehicles at the end of their lifetime).
Enthalpy	Enthalpy describes the internal energy of a system, and is used to describe energy changes or energy transfers.
EOL	End Of Life.
Eutectic impurities	Eutectic impurities have a distribution coefficient lower than 1 in a solvent. In the context of this thesis this means that at equilibrium the impurity have a higher distribution in liquid state compared to solid state in a binary solution for aluminium.
Eutectic point	The point in a phase diagram at which a single compound, composed of the present elements, forms a solid in a eutectic system.
Eutectic reaction	The reaction which takes place at the eutectic point. A single compound solidifies in another wise liquid solution at a given temperature.
Eutectic system	A eutectic system is a system which consists of various chemical compounds, and where one single compound solidifies at a lower temperature than any other composition from the same ingredients.
Extrusion	Forming of wrought products by forcing the metal through aperture dies.
Fe	Iron.

Fluxing	Refining method which uses the addition of a chemical compound to remove tramp elements.
Forging	Forming of wrought products by pressing the metal into a shaped die, the die can be either open or closed.
Gibbs free energy	Determines the direction of a reaction, the most negative reaction will take place first ($\Delta G<0$ are spontaneous). States the energy needed for the reaction to take place at a given temperature.
Inclusions	Solid particles suspended in the molten aluminium, for example oxides, they can be both metallic and non-metallic.
Ionic liquid	Organic salts in molten state, usually with a melting point < 100°C.
LCA	Life Cycle Assessment.
Mg	Magnesium.
Mn	Manganese.
New scrap	Scrap from industrial sources. Composed of leftover materials generated in production (Schlesinger, 2007), collected metal before its use phase.
Non-continuous process	The process applies one batch of scrap at a time and can be repeated on the same batch to achieve a higher purity level.
OEA	Organisation of European Aluminium Refiners and Remelters.
Old scrap	Understood as post-consumer scrap. Metal collected after its use phase.
Open-loop recycling	Various scrap is collected and recycled back into different products.
ppm	Parts per million (= 0.0001 %).
Primary metal	Metal produced from ore found in deposits of earth’s crust (Schlesinger, 2007).
Purity level	In this context used to describe the content of pure aluminium in an alloy.
Refining	The process of adjusting the alloy composition within the desired limits (Wei, 2012).
Rolling	Forming of wrought products by rolling it between two cylinders.
Secondary metal	Metal acquired from recycling (Schlesinger, 2007).
Segregation	Refining method which separates between elements in liquid and solid state.
Si	Silicon.
Skimming	Technique to remove floating matter in a liquid.
Slag	Collection of compounds removed from molten metal.
Stressor	Broader term for emissions related to a process, used to include for example depletion of raw materials, land use etc.
Tramp element	Term used to describe unwanted elements.
Wrought alloy	These alloys require a high level of purity, between 90-99.999% but most are over 95 % (Schlesinger, 2007). Wrought alloys are turned into consumer products by a solid-state process; extrusion, forging, or rolling.
wt.%	Weight %, refers to the percent content of an element in the aluminium alloy.
Zn	Zinc.

1. Introduction

1.1. Aluminium production and recycling

Aluminium is a very useful material in many areas due to properties like strength and lightweight. Other desired properties are its resistance to corrosion, its non-toxicity and as an excellent conductor of electricity and heat (Schlesinger, 2007, Boin and Bertram, 2005, Davis, 1993). These properties are mainly obtained when aluminium is alloyed with other elements, since pure aluminium is mechanically weak and soft (Engh, 1992).

When the lightweight of aluminium products are exploited in the transport sector it can help reduce fuel needs and hence reduce emissions. Aluminium is in addition very suitable for recycling both because the properties of the material are not altered when re-melted (EAA, 2004, Kahveci and Unal, 2000) and because recycling of aluminium only uses about a 5-10 % of the energy required to produce primary aluminium (Ashtari et al., 2012a, Schlesinger, 2007, EAA, 2004). A general production route for primary aluminium is illustrated in Figure 1. The first step is to mine the primary ores, which consist of bauxite. Approximately four tons of bauxite is needed to produce one ton of aluminium. The next step is the Bayer process which produces alumina. Alumina is the oxide form of aluminium, Al_2O_3. About 1600-3200kg of red mud is produced per ton of aluminium in the Bayer process (Hoberg et al., 1999). The next step is to extract pure aluminium from alumina through a reduction process. The high energy need (13-15kWh/kg (Petrucci, 2011)) to produce primary aluminium is mainly due to this step, which is step number three in Figure 1. This is due to the chemical difficulty of extracting the pure metal from its oxide form. Because aluminium is very reactive, its oxide form is very stable. Oxidation is a common refining method for metals (Nakajima et al., 2010), but it does not work for aluminium due to aluminium's strong affinity to oxygen. Therefore alternative refining methods must be applied.

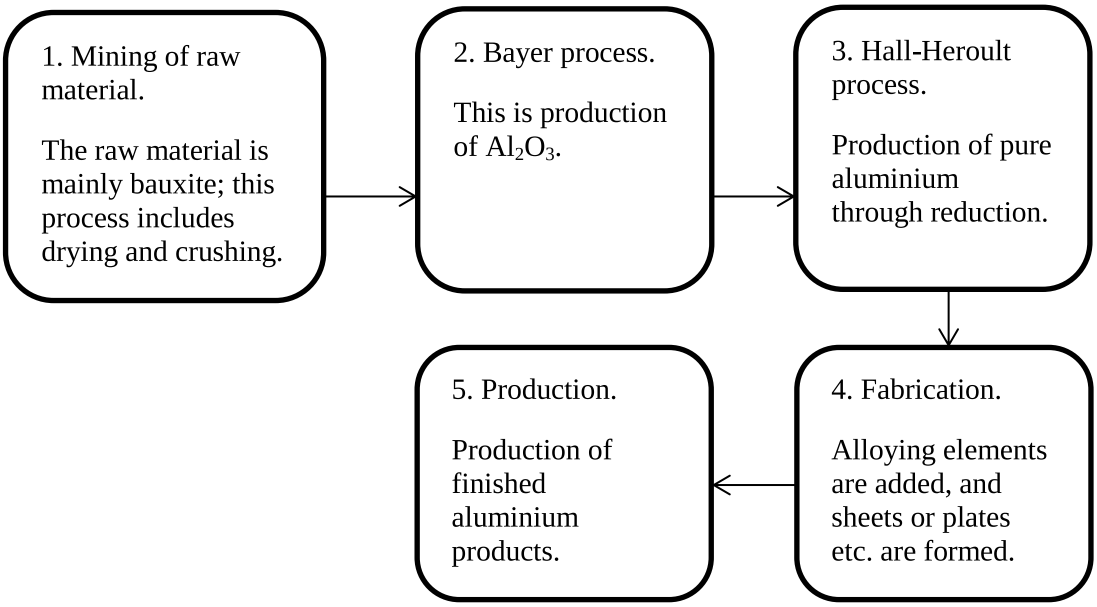

Figure 1: Production of primary aluminium products (Schlesinger, 2007).

There are two main production routes for aluminium products, and these are cast alloy production and wrought alloy production. The main differences in these are the way they are produced, the contamination limits that are allowed and the contents of alloying elements. Wrought alloys are produced from solid form through extrusion, forging or rolling. Wrought alloys typically allow fewer impurities, and are generally characterised by low content of alloying elements (EAA, 2004). A purity level of 95-99.999% is normal (Schlesinger, 2007). Cast alloys typically allow more impurities and are produced by casting directly from liquid form. Due to the low tolerance of impurities, wrought alloys are mainly produced from primary aluminium or in some cases from new scrap or old scrap diluted with primary metal. When wrought alloys are recycled they are mainly re-used in cast products. This is due to the accumulation of alloying elements and impurities when a variety of alloys are collected and melted together. This happens because various aluminium alloys and other materials are used together in the same products. As an example, a car consists of many different metals and other materials where it might be difficult to separate all the different materials and alloys in an efficient way.

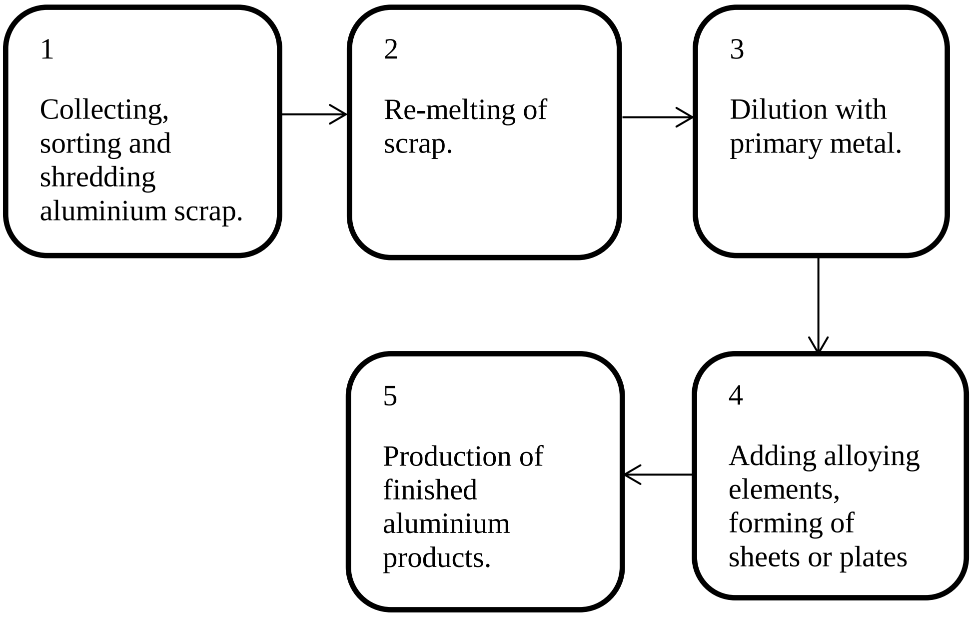

Figure 2: General production route for products containing old scrap (Schlesinger, 2007).

The current technology for recycling aluminium is called cascade recycling or downcycling. This means that the collected post-consumer scrap is melted, with some degree of dilution with primary aluminium, and re-used for products having lower purity demands (Gaustad et al., 2012, Schwarz and Wendt, 1995, Boender et al., 2002, Nakajima et al., 2010, Sillekens et al., 2000, Modaresi and Müller, 2012). This is economically preferable since it does not require much sorting of the scrap or any expensive refining processes. A general production route for aluminium products utilising post-consumer scrap is illustrated in Figure 2. Cascade recycling does not cause any problems so far, since the availability of post-consumer scrap is currently relatively low (EAA, 2004, Schlesinger, 2007). Aluminium is a relatively new material, compared to for example iron which has been widely used for a long time. Its use has rapidly increased the last decades, and the amount of aluminium in use is growing (EAA, 2004, Kevorkijan, 2002). This causes an unsaturated market for scrap aluminium, since the need for low purity cast alloys exceeds the availability of post-consumer scrap, therefore when recycling aluminium there is currently only a need to re-melt it to re-use it. Hence when it is stated that recycling of aluminium only uses a small fraction of the energy needed for primary production, it is referred to the energy needed to re-melt the aluminium scrap. Also since the melting point of aluminium is relatively low (660°C), the energy needed to recycle aluminium is low compared to primary production. The same low energy use may however not be the case if there is a desire to recycle post-consumer scrap aluminium back to products demanding high purity aluminium alloys.

It is predicted that the use of aluminium will continue to grow fast (EAA, 2004, Schlesinger, 2007), and this implies that the scrap availability will increase in the future. This means that eventually the market for downcycled scrap might saturate and excess scrap will be available. The availability of scrap is highly dependent on three factors; the amount of aluminium used in a product, the products lifetime and the collection and recovery rate of aluminium products destined for recycling (Kevorkijan, 2002). It is of interest to predict when the low-grade scrap market will saturate and the use of old scrap aluminium for other purposes might get economically and environmentally preferable. It is suggested by Modaresi and Müller (2012) that this will occur in the near future, somewhere between 2012 and 2028 depending on which dilution rate and separation methods are being used. EEA (2004) suggest that it will be economically viable to introduce separate collection of wrought alloys from cars in 2013. Rombach (2002) on the other hand suggest that it is only the volume of available scrap that will increase in the nearest decade, and not the share of available scrap compared to the production of scrap containing alloys. He does not however reflect on whether or not this will happen in a more distant future. Kevorkijan (2002) states that due to improved economy the demand for scrap will always exceed the supply, however he mention the possibility of this changing in the future, stating that there might be a point in the future where an increase in old scrap supplies will be greater than the growth of aluminium demand. The fact that the low-grade scrap market might saturate in an either near or distant future makes it interesting to develop opportunities to recycle post-consumer scrap back to products demanding higher purity standards than those currently utilising old aluminium scrap. When availability of scrap increases the price might drop, making it economically interesting to use more costly refining processes on the scrap.

There is low production of primary aluminium in Europe (4.7 million tonnes (EAA, 2004)), compared to for example China (17.8 million tonnes (World Aluminium, 2013) which is the main producer of primary aluminium. Despite the relatively low primary production, there is a large stock of aluminium containing products. At the end of their life time the aluminium contained in these products might be available for re-use. So a second incentive to develop refining opportunities for post-consumer scrap aluminium, to be used back in high purity products, can be related to security of supply of raw material. It is important to remember that since the product amount in use is increasing, one will not be able to eliminate the use of primary metal. The production of primary metal will continue to increase due to the high increase in use of aluminium in various products. But with improved recycling the increase in primary production can be minimised.

To be able to recycle post-consumer scrap back to purer aluminium alloys, there are two main routes which could be explored. The first option is to focus on sorting the aluminium post consumption. This means separating the different alloys from each other, re-melt them and use it again for the same specific product or products requiring similar alloy properties. This route might require a larger amount of available scrap to be efficient, and is easier applied to products with a shorter lifetime. The other route would be to look into refining processes to use on the post-consumer aluminium scrap. The second route is the one explored in this thesis. Because aluminium is considered a very reactive metal, traditional refining methods (such as oxidation which is used for iron) cannot be used. Therefore, refining methods which require more energy must be considered for refining of aluminium scrap.

1.2. SuPLight

This thesis is a contribution to the SuPLight project. SuPLight is an EU-funded project whose main objective is;

"Sustainable lightweight industry solutions based on wrought alloy aluminium".

The projects background is the desire to introduce more lightweight materials in the transport sector in order to reduce fuel consumption and emissions. The energy use in primary aluminium production is high compared to secondary aluminium production, so there is a further desire to be able to utilise scrap aluminium better and be able to recycle post-consumer scrap back to wrought alloys (SuPLight, 2013). SuPLight is a multidisciplinary research project which will run from 2011 to 2014. It is a collaboration between 7 countries and 11 partners, both universities and industry, and is funded by the Seventh Framework Program for Research and Technological Development (FP7). FP7 is the EU's main instrument for funding research in Europe (European Commission, 2012).

This thesis will contribute to this project by identifying possible refining options for post-consumer scrap aluminium and using life cycle assessment (LCA) to evaluate their environmental impacts.

1.3. Project aim

It is well established that collecting, sorting and melting of scrap aluminium requires much less energy than primary aluminium production. But it is not known how much the energy-needs will change when a refining process is applied to the melted post-consumer scrap to be able to meet higher purity demands.

It is technically feasible to refine any aluminium scrap composition back to high purity levels (EAA, 2004, Petrucci, 2011, Schwarz and Wendt, 1995), but this might be very energy demanding and expensive. A general goal would be to identify possible refining processes which can be preferred over primary aluminium production both from an economic and environmental perspective. The specific goal in this thesis is to take on the environmental perspective and search for solutions.

The work has been structured around three main objectives:

1) Identify and describe possible refining technologies to remove specific alloying elements present in post-consumer scrap.
2) Establish life cycle inventory for the relevant technologies.
3) Evaluate the environmental benefit of recycling post-consumer scrap back to wrought alloys, compared to primary aluminium production, using LCA.

To achieve these objectives a case scrap will be considered and different refining processes to meet the purity requirements of seven different alloy compositions will be considered. Then the different processes will be evaluated against each other and compared to both the current recycling practice and to primary aluminium production.

The post-consumer scrap composition used in this thesis is ELV (End-of-Life-Vehicles). The composition is based on earlier work for the SuPLight project presented in the report *'Metal composition levels in recycled AlMgSi alloys'* from 2012 by Hammervold et al. (2012). The alloys to be produced by the identified refining processes are based on property testing by SINTEF Raufoss Manufacturing and are found in the report *'Silicon content limitation level on formability and product performance in cast and formed* AlMgSi' from January 2013 (Gulbrandsen-Dahl et al., 2013).

The different ELV scrap cases identified by Hammervold et al. (2012) are summarised in Table 1. 'New alloy' are the suggested wt.% limit for the different alloying elements based on the composition of the ELV scrap cases. All numbers are given in wt.%.

Table 1: Case scrap composition (Hammervold et al., 2012).

Case	**Si**	**Mg**	**Fe**	**Cu**	**Mn**	**Zn**	***Purity level***
(Kirchain and Cosquer, 2007)	6.11	0.53	0.53	1.66	0.23	0.86	90.08
(Gesing, 2005)	0.53	2.52	0.46	0.20	0.53	0.29	95.47
(Zapp et al., 2002)	0.73	1.49	0.42	0.14	0.47	0.46	96.30
'New alloy'	2	1.2	0.5	0.5	1	0.2	94.6

Kirchain and Cosquer (2007) represent a study which contains future scenarios for scrap aluminium. It is not stated whether this scrap is wrought- or cast alloys or both, but it is probably a mix. Gesing (2005) and Zapp et al. (2002) present scenarios based on observations at car dismantling businesses and it is given that these are wrought alloys. It is observed that Gesing (2005) and Zapp et al. (2002) obtain numbers within the same range. Kirchain and Cosquer (2007) separate from the other two cases with larger contents of 4 out of 6 elements, and a slightly different composition. For this thesis it was decided to continue with Kirchain and Cosquers (2007) scrap composition, as this gives more elements to work with. Gesing (2005) and Zapp et al. (2002) are included for later comparison and discussion.

Table 2 displays the different alloy compositions that it is desirable to achieve after the refining processes. Alloys 1-7 are developed by SINTEF Raufoss Manufacturing and represent possible wrought alloys that could be produced using post-consumer ELV scrap. The first row contains the scrap to be refined in this thesis. All numbers are given in wt.%. Alloys 1-7 are developed based on the standardised wrought alloy AA6082, which is a commonly used wrought alloy in the transport sector (11 % in 2002 (Rombach, 2002)). Industrial pure aluminium is used as a base for the alloys, and the alloying elements are then added in different concentrations and tested. Large variations for all the elements were not tested, and due to this the limits are zero for Zn and for Cu for some of the alloys. Limits of zero are not realistic for scrap. Thus alloy 1-7 are not produced from scrap, but the idea is that these are possible alloys that *could* be produced from ELV scrap. Alloying elements that have lower contents in alloy 1-7 and criteria for ‘New alloy’ compared to the scrap are highlighted with a grey background colour. The percentage of alloying element that must be removed to meet the criteria of alloy 1-7 and ‘New alloy’ are given in braces behind the wt.% of the alloying element. The numbers are rounded to the nearest per cent.

Table 2: Finished alloy compositions. Percentage of the present alloying element to be removed is included in braces (Gulbrandsen-Dahl et al., 2013).

	Si	Mg	Fe	Cu	Mn	Zn
Scrap composition	6.11	0.53	0.53	1.66	0.23	0.86
1	1.05 (83 %)	0.81	0.25 (53 %)	0 (100 %)	0.54	0 (100 %)
2	1.98 (68 %)	0.88	0.23 (57 %)	0 (100 %)	0.59	0 (100 %)
3	3.73 (39 %)	0.84	0.23 (57 %)	0 (100 %)	0.58	0 (100 %)
4	5.51 (10 %)	0.84	0.23 (57 %)	0 (100 %)	0.58	0 (100 %)
5	1.03 (83 %)	0.82	0.66	0 (100 %)	0.51	0 (100 %)
6	1.02 (83 %)	0.75	0.25 (53 %)	0.91 (45 %)	0.54	0 (100 %)
7	1.02 (83 %)	0.79	0.65	0.88 (47 %)	0.5	0 (100 %)
New alloy'	2.00 (67 %)	1.2	0.5 (6 %)	0.5 (70 %)	1	0.2 (77 %)

The following chapter will present the theory behind this work. First the LCA methodology is presented, and then different aspects of processing secondary aluminium are explored. The LCA conducted as a part of this thesis will start from chapter 3.

2. Theory

2.1. LCA

This section gives a general description of the LCA methodology. It is based on two standards, (ISO 14040:2006, 2006) and (ISO 14044:2006, 2006) and the learning material '*Methodological issues of LCA*' by Anders Hammer Strømman for the course TEP4222 LCA at NTNU (draft of 2010) (Strømman, 2010).

LCA is a method used to identify the environmental performance of a product. It considers the entire life cycle of a product, from the extraction of raw materials through production, use, disposal and/or recycling. Different variants of the LCA methodology exist; for example only parts of the life cycle can be investigated. A cradle-to-gate analysis can be carried out, which includes the extraction of raw materials through some processing to the stage where it is ready to be used in further production or as a finished product. An example can be the production of a certain amount of metal ready to be used in the production of different products. A cradle-to-cradle or cradle-to-grave analysis, depending on decisions regarding recycling, includes the entire life cycle, including the end-of-life (EOL) treatment of a given product.

There are four phases of an LCA study;

(a) Goal and scope definition
(b) Life Cycle Inventory (LCI)
(c) Life Cycle Impact assessment (LCIA)
(d) Interpretation

Defining goal and scope: An LCA is structured around a functional unit. An example of a functional unit is 1 kg of metal ready to be used in further production, which would be a cradle-to-gate analysis, or a car used its entire life time, including EOL treatment, which would be a cradle-to-grave analysis. The functional unit can also be referred to as the external demand placed upon a given system. The first phase of the LCA consists of defining this functional unit, and deciding on the system boundaries for the study. What do we want to find the environmental impact of and within which limits. The system boundary also states which environmental impacts are included in the assessment. It is important to describe what the intention behind the LCA is, i.e. the goal of the assessment. It can for example be used to assess how environmentally friendly a specific product is, or it can be used to compare different production routes for a product.

Life cycle inventory (LCI): Inventory consists of establishing the inputs and outputs for all the different processes included within the system boundary. Inputs are typically energy, material and equipment requirements. Outputs are typically waste, emissions and/or by-products.

Life cycle impact assessment (LCIA): In the assessment phase the total output from all the included processes, due to the functional unit, is calculated. Consequently the stressors and environmental burdens from each process can be established. A stressor is a broader term for emissions, including for example fresh water use. The environmental burden related to each of the individual processes can then be summarised to evaluate the total environmental impact caused by the entire life cycle of the functional unit.

The assessment phase also includes how the different stressors contribute to different impact categories. An impact category is for example global warming potential (GWP) or human toxicity potential (HTP). A stressor can contribute to more than one impact category, and one impact category can be influenced by many stressors. To make results readable and useful the contributions from different stressors to the same impact category are converted to equivalents. For example all contributions to GWP are converted to kg CO_2-equivalents.

Interpretation: Interpretation mainly consists of discussing the results and putting them into context. But interpretation should also be conducted continuously throughout the assessment. This is illustrated in Figure 3. When interpretation is conducted continuously, the other three phases can be adjusted to enhance the quality of the assessment.

When interpreting the obtained results reflection regarding transparency, weighting and allocation should be included. Transparency is related to how decisions and assumptions have been made. It is important to identify whether or not the results are based on vague assumptions and/or imprecise parameters. Parameters can be given with an uncertainty related to them, and the uncertainty can be propagated through the system. Therefore all uncertainties must be thoroughly accounted for. It is also important to be clear on how processes etc. are defined to make results useable for others. Weighting is related to how different environmental impacts are weighted against each other. Before making recommendations based on the results it must be clear which impact category is being prioritised and why. For example fresh water use is not an issue in Norway, since availability of fresh water is high. Fresh water use may however be considered a big issue in African countries where they experience massive droughts. Allocation is related to how environmental inputs are distributed between different outputs of a process.

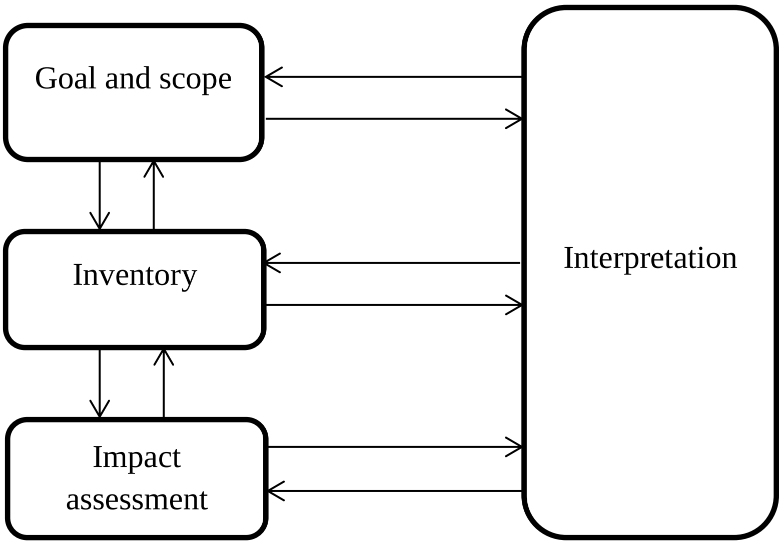

Figure 3: Principles of LCA

2.2. Alloying elements present in scrap aluminium

This section wants to provide a brief description of the alloying elements present in the scrap composition related to this thesis. This is included to supplement the description of the refining processes and the discussion of the results obtained. The aim of this section is mainly to answer these questions:

- What are the main sources of these elements?
- How do the elements affect the quality of the alloy?
- How can they be removed from the melt?

Bauxite, which is as mentioned the primary ore of aluminium, consists of approximately 50 % alumina, 10-20 % water and a various selection of other elements which are viewed as tramp elements (Tan and Khoo, 2005). These elements are mainly iron and silicon oxides. All the alloying elements present in the case scrap are found in the primary aluminium ores, but with the exception of Fe and Si they are present in negligible amounts and are thus not a target for removal during primary production.

Iron, Fe

Sources of iron in primary aluminium are mainly the minerals Fe_2O_3 and Fe_3O_4 from bauxite and through corrosion of equipment used during production, but the equipment does not provide a significant contribution. During recycling additional iron contents can occur due to co-use of steel and aluminium in products, for example in cars. It can be difficult to separate small pieces of iron from the aluminium, which results in iron being present in the aluminium melt.

Iron has detrimental impacts on the strength and ductility of aluminium alloys making it brittle and weak (Ashtari et al., 2012b, Zhang et al., 2011, Chen et al., 2011, Dewan et al., 2011). It is therefore very important to remove. There are different levels of Fe tolerated in various alloys depending on the desired qualities of that alloy. Since strength and ductility are in general important properties of aluminium alloys, Fe seems to be considered the most important impurity to remove (Dewan et al., 2011, Zhang et al., 2011). The urgency to remove Fe is mainly due to the hard A_5FeSi intermetallics that occur due to Fe containing scrap (Ashtari et al., 2012b). Ashtari et al. (2012b) clearly states that these intermetallics allow for stress concentration points which facilitate crack formation. Ashtari et al. (2012b) further states that *'controlling the Fe content in recycled aluminium alloys is crucial and strategies have to be developed to either modify the negative effects of Fe or reduce the Fe content to a predetermined acceptable range'*.

Unfortunately iron is also the most pervasive impurity element in aluminium alloys. Fe is more difficult to remove the lower the content is (Zhang et al., 2011). Many technologies for the removal of Fe from aluminium-melt have been tested during the last decades, and a selection of methods is represented here. Iron has high solubility in molten aluminium and forms intermetallic particles in solid aluminium (Davis, 1993). Iron is also considered to be a non-reactive element compared to aluminium.

Ashtari et al. (2012b) experimented with adding Mn to the melt and using a settling technique to precipitate the iron from the melt. Their theory was that adding Mn would encourage the formation of AlFeMnSi, which will result in sludge that is denser than the aluminium melt and hence sink to the bottom of the furnace. Their results together with thermodynamic calculation showed that up to 62% of the iron content could be removed. They found that as the ratio Mn/Fe increased so did the precipitation of Fe. They include the results of other studies using Mn to remove Fe. These studies report 60-70 % reduction of Fe content for melts with Fe level ranging from 0.6 to 1.2 %, and a Mn level ranging from 0.3 to 2.15 %.

Ashtari et al. (2012b) also describes other experiments on Fe removal. For example through forced filtration using ceramic foam filters that collects intermetallic particles under the filter when pushed through the melt. This might seem efficient, reporting 88% Fe removal, but difficulties arises when dealing with large amounts of aluminium, filters clog and the process cannot be made continuous.

Zhang et al. (2011) discuss methods to neutralise the detrimental effects of Fe. They have reviewed techniques to remove iron by addition of Mn, Co, Cr, Be and Sr to form intermetallic particles which can be removed by gravity sedimentation, filtration, centrifugal separation or electromagnetic (EM) separation. These are techniques that are generally used to remove inclusions in the melt. When the added elements react with Fe (or other alloying elements) they can form particles that behave similar to inclusions, therefore techniques used to remove inclusions can be applied. But there are problems related to this. The holding time for settling is high, about 180-380 minutes. The holding time is greatly reduced when a filter is applied, down to about 15-30 minutes, but then problems such as clogging of the filters occur. It is stated in their paper that Mn is the most widely used element to remove Fe. They also look into refining processes such as electrolysis, fractional crystallisation, electroslag refining and fluxing. They conclude that the three-layer electrolysis process is the most efficient way to remove Fe so far. This process will be further investigated and explained in section 2.3.2. Electrolysis.

Gao et al. (2009) studied the effect of using a $Na_2B_4O_7$ containing flux to remove Fe. They found that such a flux could reduce the iron content from 0.14 wt% to less than 0.1 wt% and achieve an optimal removal of 44 %. A holding time of 30-40 minutes is recommended, because a longer holding time will cause more oxidation loss (Gao et al., 2009).

Chen et al. (2011) studied the use of electroslag refining to remove iron impurity. In this type of refining process a flux is used both as a heat source and as a refining medium. In this case $KCL\text{-}NaCl\text{-}Na_3AlF_6$ slag containing $Na_2B_4O_7$ was used. They were able to achieve a reduction slightly above 50%.

Silicon, Si

Together with iron this is the most common element found together with aluminium in the primary aluminium ores. Si is together with Fe always present in both primary and secondary aluminium metal (Boender et al., 2002). Silicon is frequently used as an alloying element, the wt.% varies for the different alloys. Si is mainly added to increase strength, and is generally present in larger amounts in cast alloys than in wrought alloys (Rombach, 2002).

Ashtari et al. (2012a) investigated an option to remove Si from recycled aluminium alloys. They stated that Si can be removed directly from the melt during a eutectic reaction. They proposed an option to add Ca to form intermetallic particles, and remove those. They added 2% pure Ca at 675°C, reduced the temperature to 650°C, then held it for 30 minutes and then let it solidify. The prediction was that intermetallics particles, CA_2Si and $CaSi_2Al_2$, which are less dense than Al, would form and float to the top of the melt, hence reducing the Si content. It was found that this process could reduce the Si content by nearly 50 %. A problem with this method may be dispersion of particles containing Ca in the melt, which can cause trouble. Analysing the solid after this process showed that come excess Ca was present in the middle and bottom of the solidified refined metal confirming the concern.

Zhang et al. (2011), who mainly studied how to reduce the iron content, also concluded that the three layer electrolysis is effective to remove Si from the molten aluminium. Utilizing a eutectic system, Si can also be removed using fractional crystallisation. This method will be further explained in section 2.3.3. Fractional crystallisation.

Magnesium, Mg

The presence of magnesium in primary ores of aluminium is negligible, and its main source is therefore through addition as an alloying element. Consequently the amount of Mg in scrap depends on the alloy being melted and whether it is treated as an impurity or not depends on the alloy being produced. Because magnesium is more expensive than aluminium it is encouraged to use Mg containing scrap to produce alloys which requires Mg (Schlesinger, 2007).

Mg is added to improve mechanical properties, like ductility and strength (Davis, 1993), without impairing the corrosion resistance (Dewan et al., 2011). The content of Mg is often higher in wrought alloys than in cast alloys (Davis, 1993, Utigard, 1998, Rombach, 2002). This, together with the fact that Mg is expensive, is a good incentive to recycle wrought alloys back to wrought alloys. Mg is considered a reactive metal and can therefore easily be removed by fluxing (Dewan et al., 2011). Fluxing with chlorine gas is the most widely used technique to remove excess Mg from aluminium (Qian and Evans, 1998, Fjeld et al., 2006). According to an Ellingham diagram (see Appendix A), Mg can also be removed by oxidation. Oxidation can be risky due to quick formation of Al_2O_3. Electrorefining is able to remove Mg, due to the accumulation of Mg in the electrolyte (Schwarz and Wendt, 1995).

Manganese, Mn

The main source of Mn in scrap metal is due to addition as an alloying element. Its presence in primary ores is negligible. Mn is generally present in larger amounts in wrought alloys than cast alloys (Rombach, 2002). Manganese is added to increase strength in the alloy and to retard grain growth during ingot reheating. Mn can also reduce resistivity of the alloy (Davis, 1993). As mentioned Mn can be added to remove Fe (Zhang et al., 2011, Chen et al., 2011), but it can have harmful effects on the alloy in excess amounts, since it can increase the amount of insoluble microscopic particles that promote fracture (Davis, 1993). In some cases Mg is added to remove Mn (Dewan et al., 2011).

Copper, Cu

Copper is only present in primary ore in negligible amounts, and its main source is therefore due to addition as an alloying element. Copper is generally more present in cast alloys than wrought alloys (Rombach, 2002). It is mainly added to increase the strength of the alloy, both cast and wrought alloys (Schlesinger, 2007). Cu can be removed by fractional crystallisation (Boender et al., 2002).

Zinc, Zn

Zinc might be present in small amounts in the primary aluminium ores (Davis, 1993), but is mainly added to increase the strength of the alloy (Schlesinger, 2007). Zinc is the only one of the tramp elements in the case scrap that has a lower boiling point than aluminium. The boiling point of Zn is 907 °C, see Appendix A. Due to this zinc can be removed by distillation. One way to do this is a method called sweating, or selective melting. This is done in a reverberatory furnace where the temperature is stepped and held at different temperatures to easily remove materials with a lower boiling point than aluminium. One study showed that distillation could reduce zinc from 3 wt% to 0.1 wt%, which corresponds to about a 97 % removal (Gaustad et al., 2012). It can be difficult to remove Zn in small concentrations (Nakajima et al., 2010). Zinc has high solubility in solid aluminium (Davis, 1993); it is therefore difficult to remove using fractional crystallisation.

Table 3: Some of the properties related to the alloying elements present in case scrap.

	Si	**Mg**	**Fe**	**Cu**	**Mn**	**Zn**
Source of tramp element						
Present in primary ore	x	negligible	x	negligible	negligible	negligible
Added as alloying element	x	x	rarely	x	x	x
Corrosion of production-equipment			some			
Foreign metals in recycling stream			x			x
Alloying properties						
Increase strength	x	x	sometimes	x	x	x
Increase toughness				x		
Increase corrosion resistance					no effect	
Increase formability/ductility		x				
Prevent grain growth					x	
Solubility in solid			low		relatively low	high
Solubility in liquid			high			
Reduce resistivity					x	
Harmful effects						
Form particles that promote cracking			x			
Reduce toughness	x		x			
Decrease elongation				x		

2.3. Refining methods

This section describes the refining processes which were identified to be able to refine the post-consumer scrap described in Table 1 in the introduction. Fe and Si are generally the elements requiring most effort to remove. Table 2 in the introduction describes how much of each alloying element that must be removed to reach the limits set by alloy 1-7 and 'New alloy'.

Criteria that must be met by the refining processes are that they are able to remove some, or all, of the excess alloying elements present in the post-consumer scrap. It is preferable if the processes are continuous, since this more easily can be up-scaled to industrial use, and continuous processes are generally more efficient. A non-continuous process, which requires batch-wise handling, is also uneconomic since it will require more operators to handle filling and emptying the holding furnace.

2.3.1. Fluxing

Fluxing is the term used when a chemical compound is used in the treatment of molten aluminium (Davis, 1993, Utigard, 1998). A chemical compound is added to remove an impurity in one of three different ways:

1. Reacting with the tramp metal, making a denser compound which settles in the melt. This compound can be removed by gravitational settling, centrifugation or through filtration.
2. Reacting with the tramp metal, making a less dense compound which becomes a part of the slag/dross layer to be removed by skimming.
3. Increasing the fluidity/wettability of the melt, this facilitates separation of inclusions. Inclusions are generally removed by filtration, settling or in some cases centrifugation.

In general this technique is the most widely used technique to remove impurities in molten aluminium and prevent formation of Al_2O_3 by using a flux to cover the melt. The most common technologies are bubbling with chlorine gas or the use of a liquid inorganic salt layer (Utigard, 1998). Fluorides or chlorides are the preferred compounds to react with tramp metals (Davis, 1993). This is because they are very reactive elements that form stable compounds. Fluxing is generally temperature dependent. The temperature must be high enough to provide good contact and reactivity and to achieve good physical separation (Gaustad et al., 2012), but low enough to prevent formation of unwanted compounds.

Fluxing is used both in primary and secondary aluminium production. For scrap melts it is mainly used to remove Mg (Fjeld et al., 2005). In this thesis Mg is not an issue, but when looking back at Table 1 it is evident that Mg is in excess for both the other two scrap cases. It is also known that these two scrap cases consist of only wrought alloys, and that a high Mg content usually characterises wrought alloys. This results in the assumption that even though Mg removal is not essential in this case study, it might be worth including some description and inventory since it is perceived highly relevant with regards to future recycling of products containing wrought alloys, and thus might contain a larger wt.% of Mg. The two main fluxing techniques are described here.

Fluxing with Cl_2 gas

Fluxing with chlorine gas is the most common technology for removing impurities from the melt (Qian and Evans, 1998, Fjeld et al., 2006), and is especially useful for removing Mg. The chlorine gas is added to the melt through a rotating or still rod which releases the gas under the melt surface. The bubbles react with the Mg in the melt, and bring the impurities up to the surface of the melt. A simple illustration of this is viewed in Figure 4.

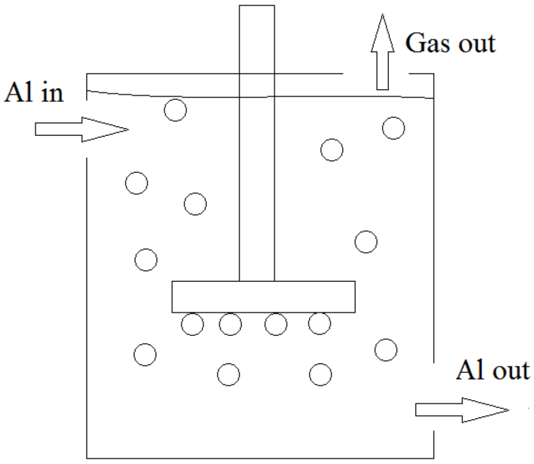

Figure 4: Fluxing with Cl_2 gas.

All metal chlorides that have a standard Gibbs free energy value more negative than $AlCl_3$ are more stable than $AlCl_3$. This means that when Cl_2 is injected to the melt containing various metals, the chlorine will preferably react with these metals. The same principle applies to fluorides. Li, Na, K, Ca, Mg and Ba all form more stable chlorides and fluorides than aluminium and can therefore be removed by Cl_2, F_2 or SF_6 (Utigard, 1998). $MgCl_2$ is a liquid at 712°C and is less dense than aluminium, hence it will become a part of a dross/slag layer at the top of the melt and Mg can then be removed from the melt by skimming.

A concern when using chlorides and fluorides is that these generate toxic gasses. Such emissions must be kept to a minimum. Emissions can be kept low provided that the temperature is kept above the melting point of $MgCl_2$ and that the Mg content is above a critical level. This level is found to be approximately 50 ppm (Utigard, 1998). The efficiency of such a process drops as the Mg content gets lower, and the removal rate is strongly dependent on the supply rate of Cl_2. The efficiency of impurity removal and control of toxic emissions also depends on gas dispersion in the melt, the residence time of the bubbles and the surface area of the bubbles. It is important to achieve sufficient contact time between the gas and the melt to get the desired reactions to take place (Qian et al., 1998). A salt layer is sometimes included when fluxing with a gas. This helps reduce emissions and keep the oxidation of aluminium to a minimum (Kevorkijan, 2010b).

Cover fluxes

Usually inorganic salts are used as cover fluxes. Normal composition of an inorganic salt layer is approximately 50/50 between NaCl and KCl (Majidi et al., 2007, Utigard, 1998). A cover flux can be used during the melting process to reduce oxidation by creating a protective layer between the melt and the air. A cover flux can also be used to remove some impurities when the elements in the flux react with the impurities making them a part of the layer on top of the melt. As mentioned above a cover flux can also be used together with fluxing with a gas to reduce oxidation and toxic gas emissions. But there are some negative impacts from using salt layers. Salts are costly in production, and the use of them results in significant amounts of salt cake production, which introduces the extra cost of salt recovery and the disposal of non-metallic inclusions in commercial and industrial landfills (Kevorkijan, 2010b).

Results from Majidi et al. (2010) conclude that the optimum fluxing temperature is 740°C for the flux to effectively remove oxides and inclusions from the melt. This temperature reduces the negative effects of a too high or too low temperature. A too high temperature will cause fume and gas formations, and a too low temperature will inhibit the desired reactions.

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Positive aspects of fluxing are that this is a very efficient way to remove magnesium (Qian and Evans, 1998), and that there are low metal losses (Utigard, 1998). The problem areas are related to addition and yield, there might be dead zones in the melt where the flux does not reach the impurities (Fjeld et al., 2005). This can cause the need of large amounts of flux to achieve an efficient reaction (Gaustad et al., 2012). Other issues are the possible release of toxic gasses when fluxing with Cl_2 gas, and some possible losses through $AlCl_3$ gas (Fjeld et al., 2005).

In the previous section where the alloying elements were described, different chemicals used to remove Fe and Si are mentioned. Ca was mentioned for Si, and Mn and $Na_2B_4O_7$ were mentioned for Fe. The use of these chemicals to remove the respective alloying elements is omitted from this case study. This is mainly due to the fact that using different chemicals which specifically remove one excess element might create a long chain of refining processes, and this might make refining of the scrap more complex. In this thesis it is more desirable to look at processes which are able to remove as many of the present alloying elements as possible. It was also stated by Zhang et al. (2011) that electrolysis was the most efficient way remove both Fe and Si, so this method is pursued next.

2.3.2. Electrolysis

Electrolysis is when an electric current is used to separate molecules by forcing an otherwise non-spontaneous process (Petrucci, 2011). When the Gibbs free energy of a process is negative ($\Delta G < 0$) the process is spontaneous, when the Gibbs free energy is positive ($\Delta G > 0$) the process absorbs energy, i.e. the process need energy to take place. This can be done by letting the reaction consume electrical energy through electrolysis. The energy losses from these types of processes are mainly through heat loss.

Three components are essential in electrolysis; two electrodes, an anode and a cathode, and the electrolyte. At the anode a reduction occurs, and at the cathode an oxidation occurs. Reduction is when an element receives electrons, and oxidation is when an element supplies electrons. Electrolyte is the solvent which transport the electrons. An electrolyte is often an inorganic salt dissolved in water (Zumdahl, 2005). These components can be altered and adjusted to optimise energy use, mass flow or efficiency. Due to this an unlimited number of possibilities exist, but with relatively small differences. A general positive attribute is that electrolysis can produce very pure aluminium (Schwarz and Wendt, 1995). Other positive aspects are that electrolysis can be applied to any scrap composition, and that there is in theory zero metal loss. Negative aspects are that electrolysis in general has high energy use, and the potential use of toxic chemicals.

For this project two main types of electrolysis have been investigated. The traditional three layer electrolysis, called Hoopes process, and the more experimental low temperature electrolysis performed on solid state aluminium scrap.

2.3.2.1. Hoopes process

As stated this is a traditional three layered process, and it is mainly used on primary aluminium to produce extremely pure aluminium. This process is structured so that it consists of three liquid layers, which are separated due to different densities of the layers. This is illustrated in Figure 5. The bottom layer consists of an aluminium copper alloy mixed with the impure aluminium, and serves as the anode. The scrap is mixed with the copper to make it denser. The top layer consist of pure aluminium and serves as the cathode. These two layers are separated by a layer of molten electrolyte (Gaustad et al., 2012). The mid layer can consist of different substances, but normally this is a layer of molten inorganic salts, usually a chloride or fluoride (Schwarz and Wendt, 1995). The idea of this process is to supply sufficient energy for the aluminium to react with the electrolyte and thus be transferred from the bottom layer up to the middle layer. When this happens, all elements more reactive than aluminium will transfer as well. Then the electrolyte is composed of components which form more stable compounds with the elements more reactive than aluminium, so only pure aluminium is able to travel up to the top layer (knowledge acquired from personal conversation with Ole S. Kjos at SINTEF Materialer og Kjemi, May 2013). This way pure Al^{3+} is transported all the way to the top layer and can be removed as very pure aluminium.

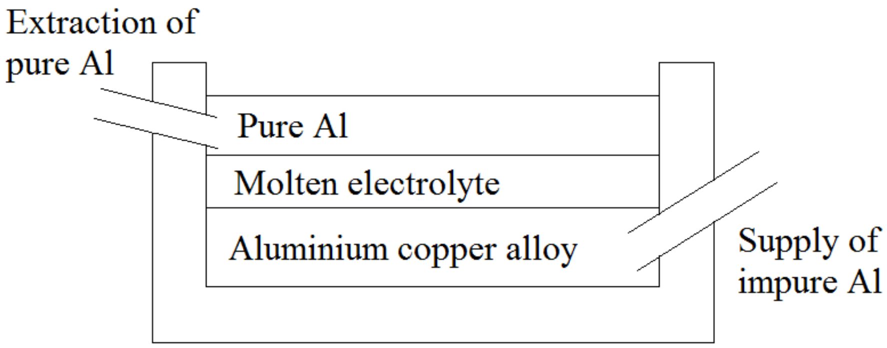

Figure 5: Principles of three-layered electrolysis.

The temperature requirement for this process is 700-900°C, and the energy use is between 17-18 kWh/kg (Kamavaram and Reddy, 2003). The process can produce aluminium with a purity of >99.07 % (Gaustad et al., 2012). It is assumed that this energy requirement include both energy to reach desired temperature, to maintain this temperature throughout the process and the actual energy consumed by the process.

Schwartz and Wendt (1995) states that this process is able to achieve a purity level comparable to primary aluminium even for scrap with a purity level as low as 70 %.

The electrolyte and copper alloy used for this process can be reused and is thus not consumed during this process. They do however require some processing to get rid of the trapped impurities before they can be reused.

Adjusted three-layered electrolysis

As mentioned the three-layered electrolysis is mainly applied to primary aluminium to produce extremely pure aluminium. Kjos et al. (2011) conducted an experiment to see if the three-layered electrolysis could be adjusted to fit aluminium scrap and possible reduce the energy requirements if the purity demands of the refined aluminium were lowered. They tested redesign of the cathode to reduce losses through voltage drop, and a new method redesigning the setup of the electrolysis. This is from a confidential report, and therefore only the energy use achieved can be included here. An energy use of respectively 8.5 and 5 kWh/kg were achieved (Kjos et al., 2011).

2.3.2.2. *Low temperature electrolysis*

This is electrolysis performed below the melting point of aluminium. When electrolysis is performed at solid state, a much lower temperature is needed and hence the energy use can be reduced. Kamavaram and Reddy (2003) conducted an experiment which only used 3 kWh per kg aluminium, and achieved a purity level of 99.89 %. This experiment utilised an ionic electrolyte. Organic salts with low melting points are called ionic liquids. Positive effects of ionic liquids compared to molten salts are lower melting point and that ionic liquids are liquids at room temperature (molten salt usually have higher melting points). The ionic liquids have a larger liquidus temperature range and high thermal and electrical conductivity which make them suitable as electrolytes. The concept used by Kamavaram and Reddy (2003) is given in Figure 6. This experiment used a temperature around 100°C, and was conducted for 2 hours. The impure aluminium serves as the anode, and pure aluminium deposits on the aluminium/copper cathode during the electrolysis. The mass flow is maintained with the help of a magnetic stirrer during the process.

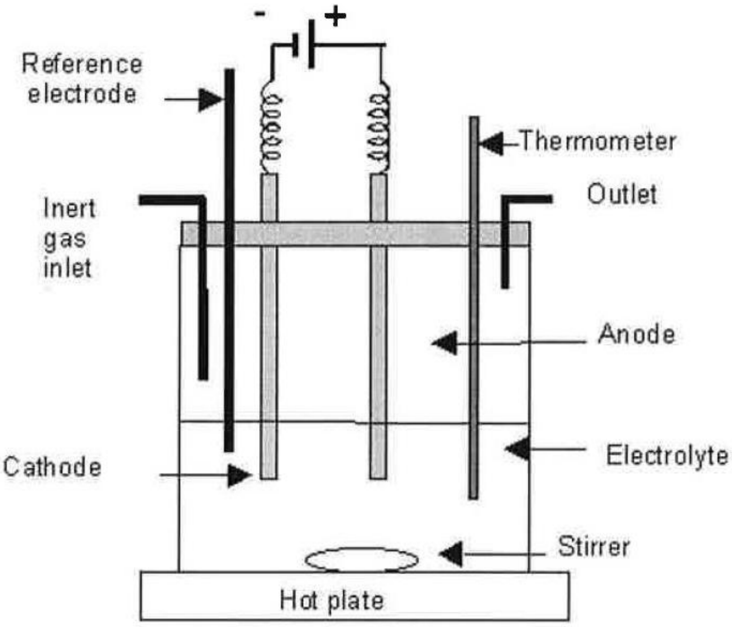

Figure 6: Low temperature electrolysis, experiment set up by Kamavaram and Reddy (2003).

The aluminium is purified through these chemical reactions;

(1) $Al_{alloy} + 7AlCl_4^- \longrightarrow 4Al_2Cl_7^- + 3e^-$
(2) $4Al_2Cl_7^- + 3e^- \longrightarrow \text{pure Al} + 7AlCl_4^-$

From the chemical equations above it is evident that no chemicals are directly consumed during this process. Elements which are more reactive than aluminium will remain in the electrolyte and the elements less reactive will remain at the anode. This follows the same concept as the three-layered electrolysis. For the scrap composition used in this thesis only magnesium is more reactive than aluminium, and will eventually contaminate the electrolyte. The excess magnesium will have to be removed at some point, but it is not found when this is expected to occur or what this removal requires.

This process has a clear main advantage of low energy use compared to the three layer electrolysis. This process is capable of removing Mn, Fe, Si, Cu, Zn, Ni and Pb (Kamavaram and Reddy, 2003). The main disadvantage related to low temperature electrolysis is the low productivity (knowledge acquired from personal communication with Mark E. Schlesinger June 2013).

2.3.3. Fractional crystallisation

This refining method is a segregation method which utilises the fact that most elements are more soluble in liquid state of a solvent than in solid state of the same solvent. The solvent in this case is aluminium. In other words; it is based on different degrees of impurity partitioning between solid and liquid state (Kahveci and Unal, 2000). This partitioning can be illustrated using a distribution coefficient which displays the ratio of an elements presence in solid state versus the elements presence in liquid state in equilibrium. The distribution coefficients for the elements related to this thesis are included in Appendix A. Fractional crystallisation is also called fractional solidification. Fractional crystallisation is, as electrolysis, developed to be used on primary aluminium to mainly remove Si and Fe to be able to produce extreme purity alloys (Sillekens et al., 2000).

Fractional crystallisation involves two main steps after the scrap has been melted (Boender et al., 2002):

1. Partial solidification of the melt.
2. Separating the solid crystals from the remaining liquid.

The principle is that the scrap is melted and then gradually cooled to partially solidify. What happens is that the remaining liquid becomes saturated with the desired compound, which in this case is aluminium, at lower temperatures. The result of this is that the excess compound will crystallise from the liquid state (Petrucci, 2011). This means that pure aluminium crystals will form in the melt. This increases the concentration of impurities in the remaining liquid and lowers the concentration of impurities in the solid state. Hence the melt is purified. Problems related to the first step are the presence of elements that have high, or equal, solubility in solid aluminium compared to liquid aluminium. These elements will either accumulate in the solid, or the concentration will stay unchanged. These elements are for example Cr, V, Zr and Ti, and they must be removed before this refining method is used. This can be done using boride formation (Gaustad et al., 2012), or filtration (Sillekens et al., 2000). In this project, the presence of neither of these elements is given, and therefore this is not accounted for in the LCA. Problems related to the second step are that pure crystals are surrounded with very impure liquid, and it can be difficult to separate the pure crystals from the impure liquid. Clear advantages with this method are no use of additional chemicals and that the main energy need occurs when melting the scrap. Some energy might be needed to keep the aluminium liquid, so the solidification can be controlled. The main disadvantage with this method is the low production yield and that the process is currently non-continuous (Zhang et al., 2011). The yield issues arise because some amount of aluminium must be left in liquid state, and are thus not part of the output of the process which is the purified aluminium. It might be possible to utilise the impure aluminium for other purposes. Parts of the less pure aluminium can be used in casted products and the very impure aluminium can be used in refining of other metals. Sillekens et al. (2002) provide an explanation of different variants of fractional crystallisation. It is distinguished between continuous and batch-wise processes. Further there is a difference in how the crystals grow in the melt, do they grow freely in the melt or as a fixed layer.

A commonly known version of this technique is the Alcoa fractional crystallisation process. This process was patented in 1980 (Dawless and Graziano, 1981). The drawing from this patent is given in Figure 7.

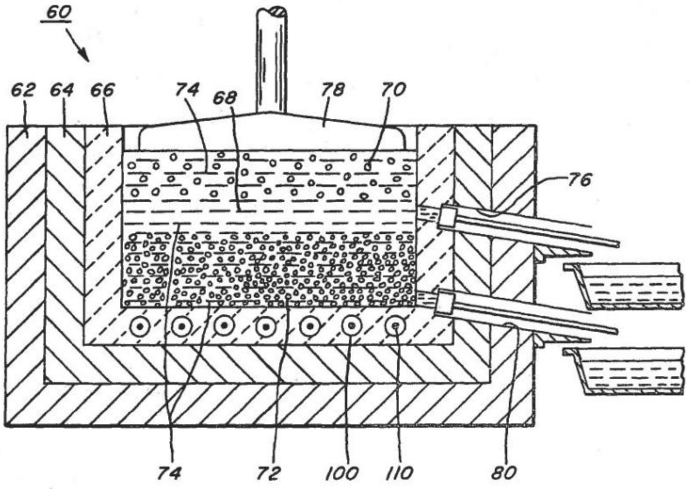

Figure 7: Fractional crystallisation, Alcoa's patented method (Dawless and Graziano, 1981).

The principle of Alcoa's process is that the aluminium melt is poured into a well-insulated apparatus called crystallisation furnace. The inner wall, *66*, must comprise of a material that cannot act as a source of contamination. It is advised to use pure aluminium for this layer. As the melt gradually cools the crystallised aluminium will sink to the bottom, and can be removed through a pipe, *80*. As time passes the purity of the crystals that are forming is reduced. To address the problems related to the pure crystals being mixed with the impure liquid, Alcoa included a second step in their patented solution. This includes an extra heating at the bottom of the furnace. When the pure crystals are re-melted at the bottom, they will tend to evacuate upwards through the layer of pure aluminium crystals. This liquefied pure aluminium will absorb some extra impurities which may have gotten trapped in the bottom layer, but more importantly it will form a purer liquid layer between the pure crystals and the impure residual melt at the top. This additional heating will increase the purification factor of the melt. This is illustrated in Figure 8. This extra heating at the bottom will increase the energy use compared to just melting the scrap and letting it gradually cool. Figure 8 displays how the purification factor is related to the yield of the process using the extra heating step. It is evident that if the yield is low the purification potential is high.

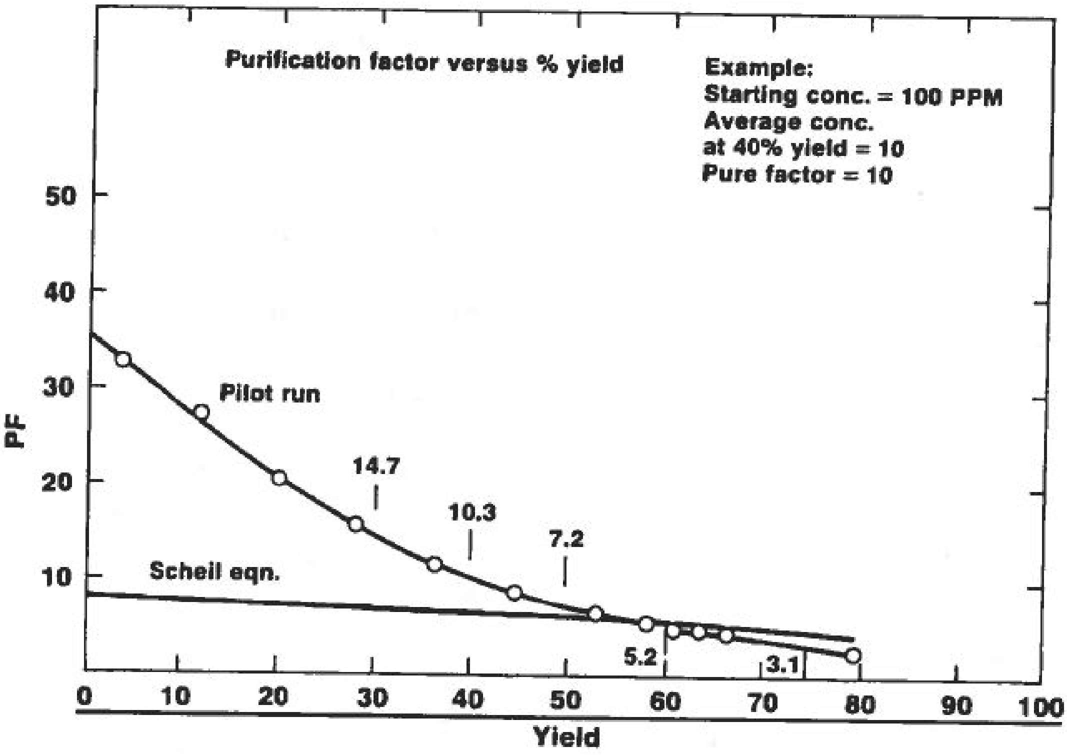

Figure 8: Purification factor of Alcoa's patent (Dawless et al., 1988).

Kahveci and Unal (2000) experimented with an extension of Alcoa's technology, so it could be used to process aluminium scrap. The desire was to be able to recycle scrap aluminium back to a similar alloy to which the scrap originated from. Kahveci and Unal (2000) state that for this method to be economically feasible, high yields of recyclable alloys must be available. In their experiment they found that the level of Si was most reduced, and that Fe had the next best removal rate. According to the distribution coefficients in Appendix A, one should expect the opposite, because Fe has a lower distribution coefficient (0.03) than Si (0.1). Kahveci and Unal (2000) explain this by the formation of intermetallics particles related to content of Fe. The removal of Cu is also observed to be less than expected, and this is also due to the formation of intermetallic particles. Formation of intermetallic particles typically occurs due to interaction between the alloying elements. For example presence of Cu and Fe might form Al_3(Fe,Cu). It can be difficult to predict the formation of intermetallic particles since distributions coefficients are based on binary solutions. Kahveci and Unals (2000) work is presented in a figure which visualises how much of each element can be expected to remain in the output from the process, depending on how much yield of the process is desired. Their figure is included in Figure 9.

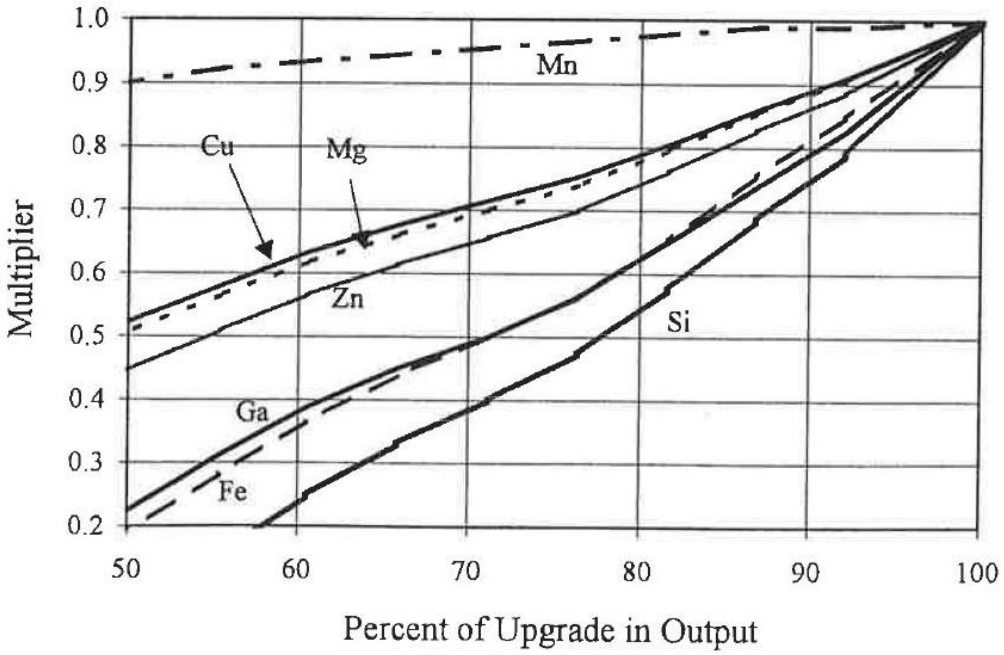

Figure 9: Presence of tramp element after fractional crystallisation (Kahveci and Unal, 2000).

The multiplier implies how many percent of the initial content concentration is present in the purified solid state. For example if the yield of the process is 60 %, the presence of Si in the output is about 25 % of the initial concentration in the melt. Looking back at Table 2, the highest removal desired for Si is 83%, resulting in a multiplier of 0.17. From Figure 9 above the multiplier is less than 0.2 for less than approximately 57 %. Therefore is it is considered plausible to expect a yield of about 50 % with respect to Si. According to Table 2 the highest removal that is required for Fe is 57 %, this is obtained within a yield less than approximately 65 %. The problems arise when looking at Cu and Zn. They both have relatively high multipliers and they both require up to 100 % removal.

Sillekens et al. (2000) state that fractional crystallisation is a possible solution to recycle various scrap aluminium on an industrial scale, and at the same time limit the energy use. But this technology has not yet left laboratory scale for scrap, and is industrially used only to produce high purity aluminium from primary aluminium. Sillekens et al. (2002) conducted a simple experiment of fractional crystallisation. After a batch of scrap was melted it was slowly cooled down and held at a temperature where solidification could occur. The results compared to theory are given in Table 5. The results given in Table 5 are based on binary systems, and it must be emphasised that for complex systems, such as real life scrap, extrapolating these results to systems with more than two components will only be indicative. It is also noticeable in Table 5 that removal of Fe and Mn requires strict process control as the available temperature interval is small.

Table 5: Results of fractional crystallisation obtained by Sillekens et al. (2002). The experimental values in this table are based on a yield of 20 %.

Element	**System**	**Max temperature range (ΔT)**	**Thermodynamic purification efficiency (η)**	**Experimental results**
Si	eutectic	83	87 %	≈ 80 %
Mg	eutectic	210	55 %	≈ 60 %
Fe	eutectic	5.5	97 %	≈ 95 %
Cu	eutectic	112	83 %	≈ 80 %
Mn	eutectic	2.5	38 %	
Zn	eutectic	279	13 %	

A further notice to Table 5 is that Mg seems to exceed the theoretical thermodynamic limit in the experimental results. Sillekens et al. (2002) suggest that this might be due to some partial evaporation of magnesium. If the desired purification level is not obtained, the process can be repeated. Sillekens et al. (2000) state that the refining performance is not dramatically affected by the initial content of alloying element, and thus the process can be repeated with a similar purifying effect. It is not confirmed anywhere how long a process like this would take, but Sillekens et al. (2002) mention that it might take several hours.

To improve the separation of purified crystals from the impure melt during fractional crystallisation, a so-called washing-column can be utilised. This is based on technology for purifying organic chemicals, but has shown promise for dealing with impure liquid adhering to the crystal surface (Sillekens et al., 2000). The technique described here, and viewed in Figure 10, was patented by Verdoes and Visscher in 2001. When applying this technique, the crystals that have formed are rinsed using a counter-currently flow of pure aluminium. In Figure 10 *1* is the inflow of metal to be treated and this is melted in vessel *2*. The melt is then fed into the wash-column *3* through line *4*. The melt passes through the filters *7* and the crystals make up a packed bottom layer *19*. A filter that can withstand the temperature of the molten metal is initially used to separate the crystals from the melt. A scraping knife *9* scrapes of the purified crystals which are transported by line *13* and re-melted in *14*. The main part of this melt is discharged via line *16*, but a small portion is recycled back into the wash-column via line *17* to wash the aluminium rich particles through a counter-current flow, thus preventing contaminants to seep through the washing layer *20* to the bottom of the column to the purified crystals *19*. The molten contaminants flow upwards in the wash-column and are discharged through line *10* (Verdoes and Visscher, 2001). It is stated in the patent that this rinsing technique is especially suitable for separating wrought alloys from cast alloys.

The use of this extra rinsing process can be used to make fractional crystallisation continuous. The idea that re-melted purified crystals can aid in the separation between crystals and impure liquid, resembles the extra melting step in the fractional crystallisation process developed by Alcoa.

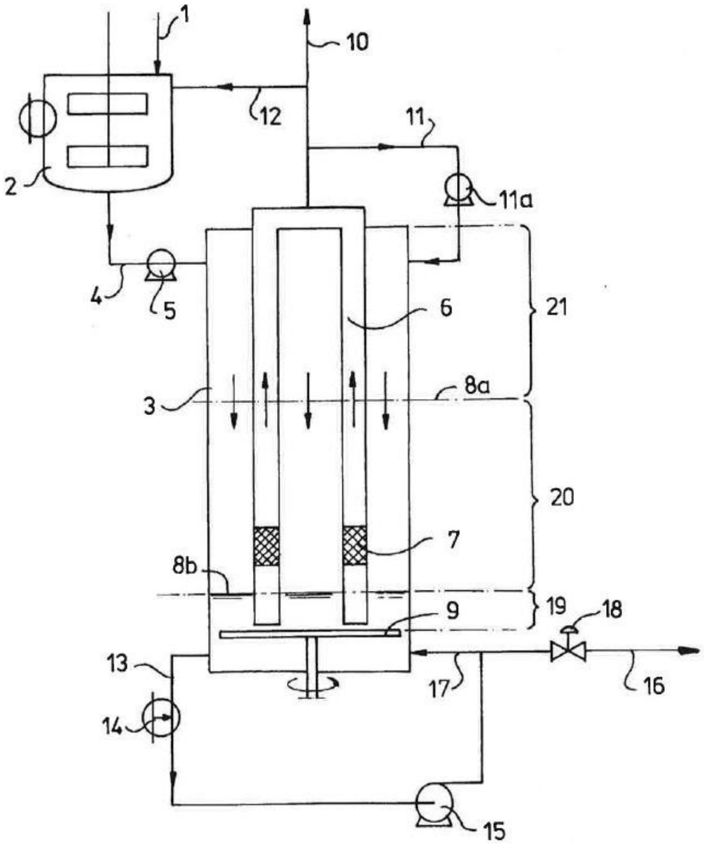

Figure 10: Principle of wash-column technique (Verdoes and Visscher, 2001).

Sillekens et al. (2000) state that the eventual implementation and success of fractional crystallisation as a refining method depend on the attainable yield and purity as well as the added value, for both product and residue, compared to the processing costs. They further state that this technology is under-explored for aluminium scrap recycling. A general restriction is that the process is slow and is of limited production capacity. Zhang et al. (2011) specifically state that this process is currently not suitable for normal foundries due to its low productivity. Sillekens et al. (2000) conclude, in agreement with Kuhvaci and Unal, that the thermodynamic potential of Fe removal is not obtained using fractional crystallisation.

2.3.4. Distillation

Distillation is a process where the substance to be purified is held at different temperatures to vaporize elements with a lower boiling point than the solvent. Gaustad et al. (2012) states that distillation holds much promise for removal of tramp elements in scrap metal, since the elements can be re-collected in a high purity state and hence be re-used as well. This refining process was mentioned related to Zn in the previous section, and this is the only element in this project that distillation is relevant for. Gaustad et al. (2012) mention a study that was able to reduce Zn content from 3wt.% down to less than 0.1wt.%. This corresponds to a 97 % removal. An issue with distillation is the presumably high production of dross due to the high temperature.

Table 6: Summary table of refining methods and which elements they are able to remove. x indicates that the refining method remove the alloying element, and (x) indicates a small removal of the element.

Refining method	**Si**	**Mg**	**Fe**	**Cu**	**Mn**	**Zn**	**Others**	**Sources**
Fluxing with Cl_2 gas		x					Ca, Li, Sr, Na, Ni, H	(Qian and Evans, 1998, Fjeld et al., 2005, Utigard, 1998)
Low temp electrolysis	x	x[1]	x	x	x	x	Pb, Ni	(Kamavaram and Reddy, 2003)
Three layered electrolysis	x	x[1]	x	x	x	x	Cr, Ti, Vr, Zr, Ga, Ni	(Dewan et al., 2011, Schwarz and Wendt, 1995, Zhang et al., 2011)
Fractional crystallisation	x	(x)	x	x	(x)	(x)	Ni, Pb, Sn, Bi,Cd	(Ashtari et al., 2012a, Boender et al., 2002, Dewan et al., 2011, Kahveci and Unal, 2000, Sillekens et al., 2002)
Distillation		(x)				x	Li, Sn, Pb, Cd, Hg, H	(Gesing et al., 2003, Nakajima et al., 2010)

[1] During electrolysis Mg is dissolved in the electrolyte and not left at the anode as the other elements in this scrap composition.

3. System definition

The system to be assessed in this LCA is a production route for secondary aluminium based on post-consumer scrap which includes a refining step. The aim is that this production route shall be comparable to the production route of primary aluminium to allow a sensible comparison between the two different production routes for aluminium.

A simplified version of an entire aluminium production system is included in Figure 11 on the next page. This figure illustrates the processes involved during aluminium production which are included in the ecoinvent database v2.1 from 2009 (Classen et al., 2009). These processes are developed closely linked to life cycle inventory work done by EAA (European Aluminium Association) with reference year 2000. These inventories were updated during 2011/2012 by EAA, but these updates are not available in ecoinvent during the work of this thesis. The processes included in Figure 11 that are related to production of secondary aluminium from old scrap are used as a basis for developing processes related to the system defined for this thesis. The process named *Aluminium, secondary, from old scrap, at plant* is the main starting point. An effort is placed on dividing this process into several sub-processes to be able to conduct a more detailed study which can provide information regarding the effects of including a refining step to the secondary production based on old scrap.

A general flow of aluminium through secondary production based on post-consumer scrap can be viewed as follows; first the metal scrap is collected, sorted and shredded. Sorting and shredding is done at for example a car dismantler. Then it is sold to remelters or refiners. Remelters are those who re-melt and reuse the aluminium as it is. Refiners apply a refining step to produce a specified alloy composition, and it is their approach that is relevant in the context of this thesis. At the refiners the molten metal is fed into a holding furnace where the alloy content and concentration is adjusted to the desired level. This adjustment can be done both by removing excess alloying elements and adding desired alloying elements. There might be some transport between the holding furnace and the foundry, where the casting takes place, and there is probably some transport between the dismantler and the refiner or remelter. Refiners often deliver the aluminium in molten state as this saves money and environmental impact (EAA, 2004).

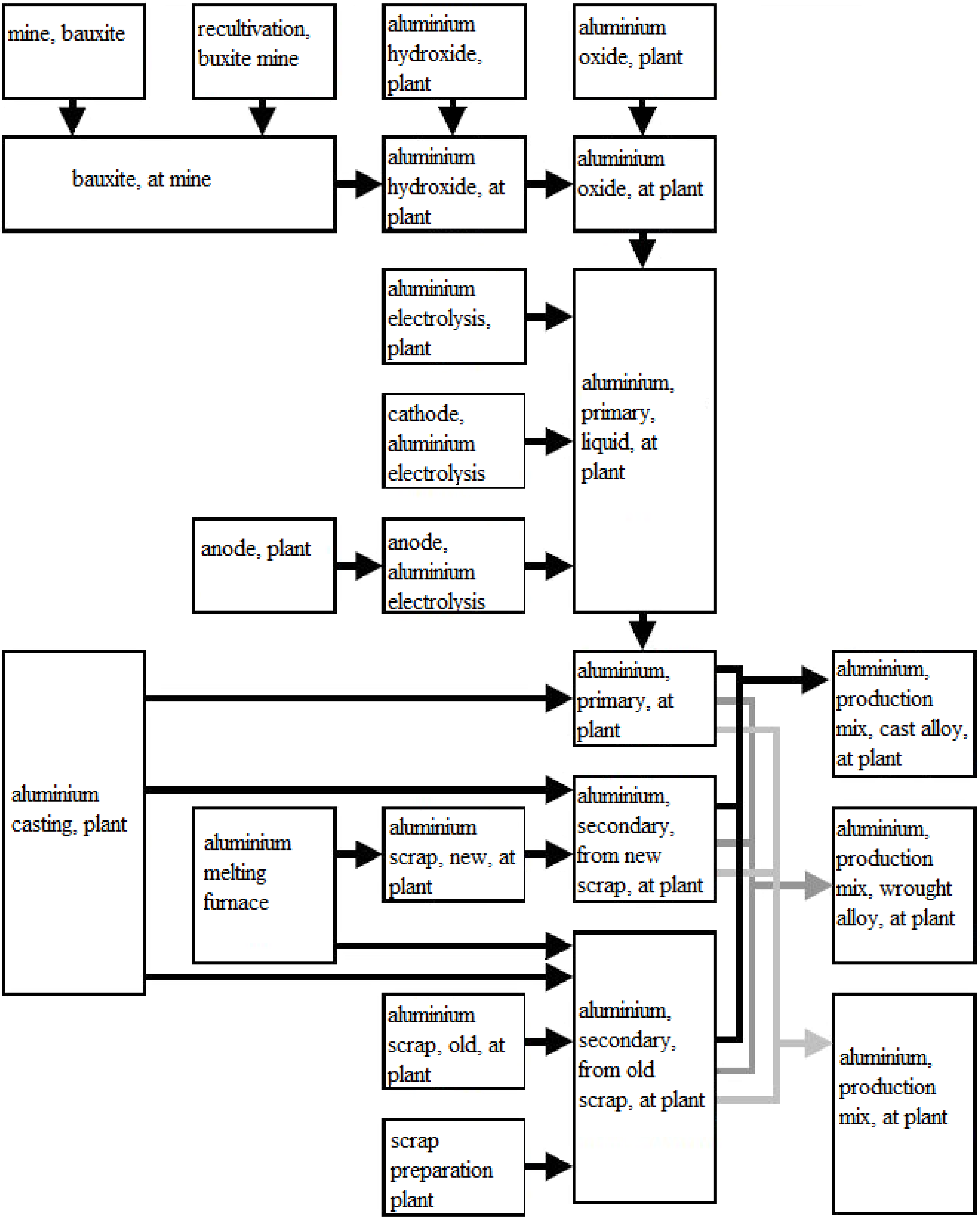

Figure 11: Simplified production system for aluminium (Classen et al., 2009).

The general idea for the system defined for this thesis is to include the processes which separate themselves from a corresponding production step in primary production. After the finished alloy exit the foundry it is considered the same whether it is produced from primary aluminium or refined post-consumer scrap. Therefore the use phase of the aluminium product is excluded from the assessment.

The collection, sorting and shredding of secondary aluminium can represent the mining of bauxite in the primary production chain. Melting and refining step of the secondary aluminium substitutes the Bayer-process and Hall-Heroult process in primary production. The casting process is considered similar for the secondary and primary production route, but is included for completeness of the system. The flowchart in Figure 12 displays the four main production steps included in the system.

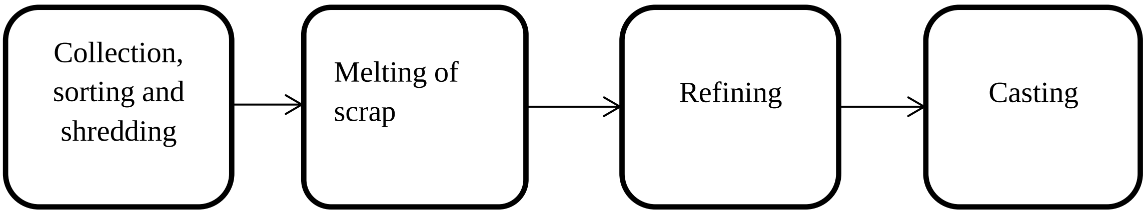

Figure 12: System flowchart.

3.1. Goal

The goal of this life cycle assessment is to investigate refining methods which makes it possible to utilise post-consumer aluminium scrap in production of aluminium products that have strict requirements to chemical purity. A further goal is to establish whether applying these methods are environmentally preferable compared to using primary aluminium to produce the same products. The investigation is based on the scrap composition given in Table 1 in the introduction and utilising the refining methods described in section 2.3. The purity limits to be reached by the refining methods are given in Table 2. A production route which is comparable to primary production is used for the refined scrap, to allow a sensible comparison to primary production.

3.2. Scope

Functional unit

The functional unit is set to be 1 kg of refined aluminium, with a content of alloying elements within the limits set by alloys 1-7 in Table 2.

System boundaries

The system boundaries are set to include the four processes displayed in the flowchart in Figure 12. The refining step is the only step that is varied during the assessment. The use of aluminium after casting is excluded. The impact categories included are given by the method used when analysing the system. The method used in this thesis is mainly based on ILCD 2011 (International reference Life Cycle Data system), a midpoint method released by the European commission joint research centre (European Commission, 2010). This method is based on LCA in a European context and includes 16 midpoint impact categories. In addition the cumulative energy demand (CED) is accounted for in the applied method. This includes 6 impact categories. The method used is based on requirements from the SuPLight project.

Included impact categories:

ILCD:

1. Climate change: Global Warming Potential (GWP)
2. Ozone depletion: Ozone Depletion Potential (ODP)
3. Human toxicity, cancer effects
4. Human toxicity, non-cancer effects
5. Particulate matter
6. Ionizing radiation HH (Human Health)
7. Ionizing radiation E (Ecosystems)
8. Photochemical ozone formation
9. Acidification
10. Terrestrial eutrophication
11. Freshwater eutrophication
12. Marine eutrophication
13. Freshwater ecotoxicity
14. Land use
15. Water resource depletion: Freshwater scarcity
16. Mineral, fossil & renewable resource depletion

CED:

17. Non-renewable, fossil
18. Renewable, water
19. Renewable, wind, solar, geothermal
20. Renewable, biomass
21. Non-renewable, biomass
22. Non-renewable, nuclear

3.3. Scenario descriptions

To do this assessment six different production scenarios have been developed. Each of these scenarios utilise different refining methods to produce aluminium which are within the purity limits set by alloy 1-7 in Table 2. All scenarios are based on the general flowchart in Figure 12. The scrap is collected, sorted and shredded using a standardised method which will be further explained in chapter 4. The scrap is melted and refined according to the applied refining method in each scenario. The casting is the same for all scenarios.

Scenario 1a: Hoopes process as refining method

This scenario involves the traditional three-layered electrolysis, known as Hoopes process, as the refining method. After shredding the scrap is melted and poured into a holding furnace appropriate for a three layered electrolysis. It is given in section 2.3.2.1. that a temperature of 700-900°C is required for this process. It is also given that the process is expected to remove any unwanted alloying elements. Therefore when applying this refining technology it is assumed that all the excess alloying elements are removed. This means that some negligible amounts are possibly still present in the purified aluminium, but this is assumed to be well within the limits set by alloys 1-7 in Table 2.

Scenario 1b: Adjusted three-layered electrolysis as refining method

This is basically the same as the previous scenario. The difference is the low energy use during electrolysis compared to the previous scenario. As described in section 2.3.2.1. Kjos et al. (2011) experimented with the effects of lowering purity demands and adjusting the set up. They ended up with a highly reduced energy need. Since the purity demand is decreased, it is not expected here that all present alloying elements are reduced as much as for the previous scenario, but since the results of the experiment by Kjos et al. (2011) where satisfactory, it is assumed that the limits set in Table 2 where met.

Scenario 2: Low temperature electrolysis as refining method

The concept of this scenario is very similar to the two previous scenarios. The main difference is that a much lower temperature is needed (75-120°C), hence the energy requirements are lowered. The low temperature implies that the aluminium is in solid state. Due to this the melting step in Figure 12 is omitted from this scenario. From section 2.3.2.2 it is understood that aluminium and more reactive elements are dissolved in the electrolyte, and that pure aluminium then deposits on the cathode. The less reactive elements are left at the anode. Based on this, it is assumed that the purified aluminium meet all the limits set by alloy 1-7 in Table 2.

Scenario 3a and 3b: Fractional crystallisation as refining method

These scenarios utilises a different refining approach than the previous scenarios. The scrap is melted and poured into appropriate apparatus to be gradually cooled. For this refining method it can be seen in section 2.3.3. that not all alloying elements can be removed with the same rate of success. It is evident in Figures 8 and 9 that the desired yield of fractional crystallisation is very closely linked with the achieved removal rate for the various alloying elements. If a yield of 50 % is assumed, as discussed in section 2.3.3., Zn and Cu are the limiting factors. Zn can be removed by distillation. To be able to remove Zn it can be assumed that after the melting process an additional distillation process takes place. The melt is heated to 900°C to remove 97 % of the present Zn, as according to Gaustad et al. (2012). Table 2 indicated that a 100 % removal is desired, but it is considered unrealistic to achieve this and that 97 % removal is assumed to be satisfactory.

According to Figure 9 the removal of Cu is the limiting factor if Zn is already removed by distillation. The desired removal range for Cu is 45-100 %. If the yield is set to 50 %, a removal of Cu can be considered close to 50 % which barely reaches the lowest removal requirement. The desired removal of Si and Fe are met with the assumption of a yield of 50 % according to Figure 9, and as discussed in section 2.3.3. Since the removal of Cu is not sufficient for alloys 1-5 with fractional crystallisation alone, dilution is required to meet the purity limits. Dilution will also reduce the excess Zn concentration. Based on this, two variants of this scenario are explored. The first version, 3a, targets alloy 1-5. For alloy 1-5, which require a 100 % removal of both Cu and Zn, it is assumed that 50 % is removed during fractional crystallisation and that dilution with primary aluminium is required to meet the purity limits related to these elements. The second version, 3b, targets alloy 6 and 7 which require a removal of Cu of 45 and 47 % respectively. It is assumed for this scenario that the excess Cu is removed by fractional crystallisation alone, and that Zn is removed by introducing an extra step of distillation.

Scenario 4: Fluxing with Cl_2 – gas as refining method

This is not directly a complete refining scenario. Fluxing with Cl_2 gas is mainly used to remove excess Mg. Since the scrap composition related to this thesis does not require removal of Mg, this scenario is considered incomplete since the used refining method does not remove any of the other tramp elements. Due to the presumed importance of this method, as discussed in section 2.3.1., it is included as an alternative scenario. In this scenario the scrap is melted, and then heated to the optimum fluxing temperature (740 °C). Then fluxing takes place. Based on this it is understood that this scenario will not refine the case scrap to meet any alloy criteria set in Table 2. A scenario like this might seem strange to include, but it might provide some interesting results that can be useful when considering other scrap compositions, especially scrap which is solely based on wrought alloys.

4. Inventory

The inventory is mainly based in the theory presented in chapter 2 together with a series of assumptions which will be explained continuously as the inventory is presented. The inventory for the three constant processes in Figure 12 will be presented first, then the inventory for the refining processes will follow in the order they were presented in section 2.3. The last section contains the inventory related to each of the production scenarios presented in section 3.3. The inventory is a mix of processes developed for this thesis and existing processes in the ecoinvent database. Process names are written in *italics* and element names are written as normal text.

The electricity mix used throughout the assessment is a European electricity production mix. The process used is called *European mix, electricity for SuPLight.* Backwards tracing of this process reveals that this contains roughly a mix of 28 % Czech production, 11 % Hungarian production, 51 % Polish production and 10 % Slovak production.

4.1. Collection, sorting and shredding of post-consumer ELV scrap

The collection and sorting process is a part of earlier work related to the SuPLight project, and used as found. The inputs to this process are different sorting methods and a material mix, and the outputs are aluminium scrap and various waste disposals.

4.2. Melting of scrap

As mentioned before most of the energy from the Hall-Heroult process in the primary production is stored in the metal, so the energy needed to melt the scrap is low compared to energy needed during primary production. The energy needed to melt scrap is, as mentioned, normally the energy need referred to when recycling of aluminium is compared to primary production. To be able to see the difference when a refining process is included it is perceived central to include this process as accurate as possible.

There are a variety of ways to melt scrap aluminium. The most widely used technique in industry is different types of combustion furnaces, which utilise fossil fuels, normally natural gas is used. But there might some changes occurring in the industry. Kevorkijan (2002) mention that melting of scrap has undergone great improvements, and that melting furnaces currently being used are more efficient and environmentally sound. Schlesinger (2007) suggests that induction furnaces are the more environmentally friendly choice. An induction furnace was also used in some of the experiments investigated for this assessment (Kahveci and Unal, 2000, Ashtari et al., 2012b). Gao et al. (2009) applied a resistance furnace, which is also an electric furnace, but a different technology. When melting scrap aluminium a salt layer can be applied. EEA (2004) state that approximately 0.3-0.5 kg/kg Al is needed, but that this salt can be reused. The salt is mainly used to prevent losses through oxidation of the aluminium, and thus provide a better quality of the melt. Without the salt layer one can expect losses about 2-3 % with a multichamber furnace (Kevorkijan, 2010b). Kevorkijan (2010b) states that a mix of NaCl and KCl is a common melting additive but that the use of salts is a costly affair. Salts are generally expensive, and the energy needed to recycle them is high. It is also expensive to dispose of the salt residues, due to them being harmful to nature. Due to the extra costs Kevorkijan (2010b) recommends salt free melting for scrap containing less than 10 % organic impurities.

Based on all the findings mentioned above, an induction furnace without a salt layer was chosen as the applied technology. Inventory for the salt layer is included as an alternative, since it appears to be about 50/50 distribution of who uses it and who does not. A salt layer is mainly used for high purity production. Even though this is not high purity production inventory has been included because it is interesting to investigate how large of an effect the salt layer will have on the environmental impacts. Unfortunately it is not stated anywhere what is required to recycle the salt or how many times it can be expected to be recycled.

Unfortunately none of the studies investigated mention the energy needed to melt scrap. This may be due to many factors, maybe because this is not the focus of any of the studies, or that the energy need might vary a lot. Variations might be due to the large variety in scrap compositions, whether the scrap is pre-heated or not and which technology is applied. It is clearly established though that the energy needed to recycle aluminium is about 5-10 % of the energy needed for primary production, and that this is mainly related to the melting of the scrap. 5-10 % of 10-15kWh/kg aluminium results in an energy need of 0.5-1.5 kWh per kg aluminium for melting of aluminium scrap. Energy calculations, included in Appendix B, resulted in an energy use of 2.24 kWh/kg to melt pure aluminium when the initial temperature of the aluminium is 298 K (25°C). When aluminium is alloyed with other elements, and contains various inclusions, the melting point is expected to decrease since the space between the present atoms and molecules will increase. EU best practise for melting of scrap is 0.5 kWh/kg (knowledge acquired from personal communication with Anne Kvithyld at SINTEF Materialer og kjemi May 2013). Based on the combination of the information above the energy needed to melt the case scrap is assumed to be 0.7 kWh/kg. This number might be lower for industry utilising very efficient combustion furnaces, and higher for low scale laboratory experiments using electric furnaces.

For this process infrastructure is included. This is based on the ecoinvent process *Aluminium melting furnace/RER/ I U*. This process is based on a medium sized plant, and assumes an output of 10,000 tons of aluminium annually and a lifetime of 50 years. This result in an input of $1/(10{,}000 \text{ ton/year} \cdot 1000 \text{ kg/ton} \cdot 50 \text{ years}) = 2 \cdot 10^{-9}$ p/ kg aluminium. The unit p represents the factory, including construction and demolition. No economic aspects of the infrastructure are accounted for, since this is solely an environmental assessment.

The main output from this process is 1 kg of melted aluminium scrap. Other outputs are two types of waste disposal. As mentioned in the system description in chapter 3, the development of processes for this assessment is largely based on existing ecoinvent processes which are developed based on life cycle inventory (LCI) work done by EAA. The two types of waste disposal included are based on the process *Aluminium, secondary, from old scrap, at plant/RER U*. This process represents an average secondary production route for old scrap and includes melting, alloying and casting (Classen et al., 2009). The waste treatments assumed to be related to the melting of scrap are included. This is based on the assumption of some solid waste related to losses in the melting process.

Table 7: Inventory for the created melting process; *Melt aluminium scrap, induction furnace.*

Input	**Amount**	**Unit**	**Source**
European mix, electricity for SuPLight	0.7	kWh/kg Al	Calculations and assumptions
Aluminium melting furnace/RER/I U	$2 \cdot 10^{-9}$	p/ kg Al	Ecoinvent process
Output			
Melted aluminium scrap	1	kg	
Disposal, filter dust Al electrolysis, 0% water, to residual material landfill/CH U	0.00963	kg/kg Al	(Classen et al., 2009)
Disposal, municipal solid waste, 22.9%, to municipal incineration/CH U	0.00008	kg/kg Al	(Classen et al., 2009)

There is also an alternative melting process created which include a salt layer. This is modelled the same way as the inventory presented in Table 8, but also includes the chemicals needed for the salt layer, and extra disposal of hazardous waste and some chlorine emissions to air. This is displayed in Table 9. It is assumed that 0.5 kg of salt is needed per of aluminium and that the composition is 50 % KCl and 50 % NaCl. These assumptions are vaguely based on Kevorkijan (2010b), Schlesinger (2007) and EAA (2004). Since no process for KCl is included in ecoinvent, a process for potassium perchlorate, $KClO_4$, is used as a replacement. The process *Sodium chlorine, powder, at plant/RER U*, is also used by the before mentioned process *Aluminium, secondary, from old scrap at plant.*

Table 8: Inventory for the created melting process which includes a salt layer; *Melt aluminium scrap, induction furnace, with salt layer.*

Input	**Amount**	**Unit**	**Source**
European mix, electricity for SuPLight	0.7	kWh/kg Al	Calculations and assumptions
Aluminium melting furnace/RER/I U	$2 \cdot 10^{-9}$	p/ kg Al	Ecoinvent process
Potassium perchlorate, at plant/GLO U	0.25	kg/kg Al	
Sodium chlorine, powder, at plant/RER U	0.25	kg/kg Al	
Outputs			
Melted aluminium scrap	1	kg	
Chlorine	$4.9 \cdot 10^{-7}$	kg/kg Al	(Classen et al., 2009)
Disposal, filter dust Al electrolysis, 0% water, to residual material landfill/CH U	0.00963	kg/kg Al	(Classen et al., 2009)
Disposal, municipal solid waste, 22.9%, to municipal incineration/CH U	0.00008	kg/kg Al	(Classen et al., 2009)
Disposal, hazardous waste, 0% water, to underground deposit/DE U	0.0145	kg/kg Al	(Classen et al., 2009)

4.3. Casting

This process has few requirements. The molten alloyed aluminium does not require any energy or resources to solidify, just appropriate equipment, time and being present in a room with a temperature lower than its melting point.

ecoinvent has an infrastructure process which can represent the secondary casting plant; *Aluminium casting, plant/RER/I U*. This process is based on a medium sized plant with an annual output of 130,000 tons of aluminium, and a lifetime of 50 years. This results in an input of infrastructure of $1/(130{,}000 \text{ ton/year} \cdot 1000 \text{ kg/ton} \cdot 50 \text{ years}) = 1.54 \cdot 10^{-10}$ / kg aluminium. In addition the operation of the metal factory is accounted for in this process. This involves general energy related to for example lighting and tap water use. It is stated in the description of this process that 1 kg of the process relates to 1 kg of metal output.

Losses from this process might be some cut-offs after casting, but no values for this are found and therefore zero loss is assumed.

Table 9: Inventory for the created casting process; *Secondary casting.*

Input	**Amount**	**Unit**	**Source**
Aluminium casting, plant/RER/I U	$1.54 \cdot 10^{-10}$	p/ kg Al	(Classen et al., 2009)
Metal working factory operation, average heat energy/ RER U	1	-	Ecoinvent process
Output			
Refined and casted aluminium	1	kg	

4.3. Refining processes

Since aluminium is a relatively expensive metal there is a general goal in the industry to minimize losses, so in theory there are no direct metal losses. But this does not mean that all the aluminium entering the refining process as melted scrap can be re-collected as pure aluminium. The amount of pure aluminium which can be extracted from each process depends on which refining method is used and is given as the yield of the process.

Fluxing with Cl_2 gas

This refining method is not directly relevant for the scrap composition related to this thesis, but it is included as described in the alternative scenario 4 as described in section 3.3. It is assumed that fluxing at 740°C can take place in the melting furnace; therefore no extra infrastructure is included. The chlorine requirement is based on Utigard (1998), and the energy requirement is based on calculations included in Appendix B. They are based on the energy needed to obtain a temperature of 740°C in the melt which is already heated to the melting point, 660°C, during the melting process.

Table 10: Inventory for the created fluxing process; *Fluxing with Cl2 gas, aluminium melt.*

Input	**Amount**	**Unit**	**Source**
Chlorine, liquid, production mix, at plant/RER U	30/1000 = 0.03	kg/kg Al	(Utigard, 1998)
European electricity mix, electricity for SuPLight	1.01		Calculation, see Appendix B
Output			
Refining related to 1 kg of aluminium	1	kg	

Hoopes process

The temperature requirements for this processes is 700-900°C (Kamavaram and Reddy, 2003). For the three layered electrolysis two options have been explored. The “traditional” energy need, which is the energy required for this process assumed in general. The problem is that this energy need is based on production of very high purity alloys, which is not the desire in this thesis. Therefore an option, using the energy number suggested by Kjos et al. (2011) mentioned in section 2.3.2.1., is also explored. Since their experiment was structured around how this type of refining process can be utilised on scrap to produce less pure alloys than those traditionally produced from electrolysis, it seems highly relevant to include in this thesis.

In section 2.3.2.1 it is stated that this process requires and anode, cathode and electrolyte. The cathode is the pure aluminium, so this does not require any input. The anode is often based on a copper alloy mixed with the scrap and the electrolyte is often inorganic salts (similar to those used as cover fluxes during melting). Unfortunately it was not found which amounts of anode and electrolyte required for such a process. But it is known that these chemicals can be re-used after some cleaning process has taken place. It is not known what these cleaning processes require, or how many times the chemicals can be assumed to be re-used. Due to this, the demand for electrolyte and anode are omitted from this assessment.

Ecoinvent has an infrastructure process which can represent the electrolysis plant; *Aluminium electrolysis, plant/RER/I U*. This process is based on a medium sized plant with an annual output of 130,000 tons of aluminium, and a lifetime of 50 years. This results in an input of infrastructure of $1/(130{,}000 \text{ ton/year} \cdot 1000 \text{ kg/ton} \cdot 50 \text{ years}) = 1.54 \cdot 10^{-10}$ / kg aluminium. Disposal of dross from the electrolysis is based on inventory created for electrolysis during primary aluminium production by Classen et al. (2009).

Inventory for the adjusted three-layered electrolysis is identical to the inventory for Hoopes process, except the lowered energy use.

Table 11: Inventory for the created three-layered electrolysis based on Hoopes process; *Three layer electrolysis (Hoopes process), old aluminium scrap.*

Input	**Amount**	**Unit**	**Source**
European mix, electricity for SuPLight	17	kWh/kg Al	(Gaustad et al., 2012)
Aluminium electrolysis, plant/RER/I U	$1.54 \cdot 10^{-10}$	p/ kg Al	(Classen et al., 2009)
Outputs			
Refining related to 1 kg of aluminium	1	kg	
Disposal, dross from Al electrolysis, 0% water, to residual material landfill/CH U	0.00011	kg/kg Al	(Classen et al., 2009)

Table 12: Inventory for the created three-layered electrolysis adjusted for scrap; *Three layer electrolysis, for scrap, old aluminium scrap.*

Input	**Amount**	**Unit**	**Source**
European mix, electricity for SuPLight	6.5	kWh/kg Al	(Gaustad et al., 2012)
Aluminium electrolysis, plant/RER/I U	$1.54 \cdot 10^{-10}$	p/ kg Al	(Classen et al., 2009)
Outputs			
Refining related to 1 kg of aluminium	1	kg	
Disposal, dross from Al electrolysis, 0% water, to residual material landfill/CH U	0.00011	kg/kg Al	(Classen et al., 2009)

Low temperature electrolysis

Kamavaram and Reddy (2003) found the energy use to be approximately 3 kWh/kg Al for this type of process. As stated in section 2.3.2.2. Low temperature electrolysis, this process does not consume any chemicals. The same infrastructure and dross disposal used for the three-layered electrolysis is included.

Table 13: Inventory for the created low temperature electrolysis process; *Low temperature electrolysis, old aluminium scrap.*

Input	**Amount**	**Unit**	**Source**
European mix, electricity for SuPLight	3	kWh/kg Al	(Kamavaram and Reddy, 2003)
Aluminium electrolysis, plant/RER/I U	$1.54 \cdot 10^{-10}$	p/ kg Al	(Classen et al., 2009)
Outputs			
Refining related to 1 kg of aluminium	1	kg	
Disposal, dross from Al electrolysis, 0% water, to residual material landfill/CH U	0.00011	kg/kg Al	(Classen et al., 2009)

Fractional crystallisation

As explained in the scenario description in section 3.3., fractional crystallisation does not require any additional chemical. The energy requirements related to this process is to keep the metal liquid to be able to control the solidification. For this an electricity use of 0.05 kWh/h is assumed (knowledge acquired from personal communication with Anne Kvithyld at SINTEF Materialer og kjemi May 2013). It was difficult to establish how long a process like this might take, so a vague assumption of 5 hours was used. This is based on a statement from Sillekens et al. (2002) that a process like this might take several hours. It was also difficult to establish what kind of infrastructure is needed for such a process to take place. It is assumed that it is too complex to take place in the melting furnace and therefore some very general infrastructure is accounted for using the ecoinvent process *Metal working factory/RER/ I U.* The description of this process state that $4.85 \cdot 10^{-10}$ units of this process is needed to produce 1 kg of final output. This is a very vague assumption, but it is assumed that the infrastructure will not account for a large share of the total impacts and therefore a very general process can be used.

Table 14: Inventory for the created process representing fractional crystallisation; *Fractional crystallization, old aluminium scrap.*

Input	**Amount**	**Unit**	**Source**
European mix, electricity for SuPLight	0.05·5h	kWh/h/kg Al	Assumption
Metal working factory/RER/I U	$4.85 \cdot 10^{-10}$	p/ kg Al	Assumption
Output			
Refining related to 1 kg of aluminium	1	kg	

Distillation

The requirements for distillation are assumed to be the extra energy needed to reach the high temperature (900°C), and infrastructure. The energy needed to reach the desired temperature is based on calculations included in Appendix B. This is based on the assumption of energy required to heat liquid pure aluminium from 660°C up to 900°C. Due to the high temperature needed it is assumed that distillation cannot take place in the melting furnace and therefore additional infrastructure is needed. The required infrastructure is based on the same assumptions as the infrastructure included for fractional crystallisation.

Table 15: Inventory for the created distillation process; *Distillation, 900C, aluminium melt, old scrap.*

Input	**Amount**	**Unit**	**Source**
European mix, electricity for SuPLight	1.54	kWh/kg Al	Calculation, see Appendix B.
Metal working factory/RER/I U	$4.85 \cdot 10^{-10}$	p/ kg Al	Assumption.
Output			
Refining related to 1 kg of aluminium	1	kg	

4.4 Production scenarios

Inventory for the six different production scenarios for refining of old aluminium scrap is included here.

Table 16: Inventory for scenario 1a – Hoopes process as refining method; *Refined aluminium from old scrap, three-layer electrolysis (Hoopes process).*

Input	**Amount**	**Unit**	**Comment**
Shredding and sorting, Aluminium	1.07	kg	
Melt aluminium scrap, induction furnace	1.05/0.98 = 1.07	kg	Assume 2 % loss from melting process.
Secondary casting	1	kg	
Three layer electrolysis (Hoopes process), old aluminium scrap	1/0.95 = 1.05	kg	Assume 95 % yield from electrolysis.
Output			
Refined aluminium	1	kg	

Table 17: Inventory for scenario 1b – adjusted three-layered electrolysis as refining method; *Refined aluminium from old scrap, three-layer electrolysis (for scrap)*

Input	**Amount**	**Unit**	**Comment**
Shredding and sorting, Aluminium	1.07	kg	
Melt aluminium scrap, induction furnace	1.05/0.98 = 1.07	kg	Assume 2 % loss from melting process.
Secondary casting	1	kg	
Three layer electrolysis, for scrap, old aluminium scrap	1/0.95 = 1.05	kg	Assume 95 % yield from process.
Output			
Refined aluminium	1	kg	

Table 18: Inventory for scenario 2 – low temperature electrolysis as refining method; *Refined aluminium from old scrap, low temperature electrolysis.*

Input	**Amount**	**Unit**	**Comment**
Shredding and sorting, Aluminium	1.07	kg	
Melt aluminium scrap, induction furnace	1.05/0.98 = 1.07	kg	Assume 2 % loss from melting process.
Secondary casting	1	kg	
Three layer electrolysis, for scrap, old aluminium scrap	1/0.95 = 1.05	kg	Assume 95 % yield from process.
Output			
Refined aluminium	1	kg	

Table 19: Inventory for scenario 3a – fractional crystallisation and dilution as refining method; *Refined aluminium from old scrap, fractional crystallization, with dilution.*

Input	**Amount**	**Unit**	**Comment**
Shredding and sorting, Aluminium	0.408	kg	
Melt aluminium scrap, induction furnace	0.4/0.98 = 0.408	kg	Assume 2 % loss from melting process.
Secondary casting	1	kg	
Fractional crystallisation, melted old aluminium scrap	0.4	kg	Assume a 50 % yield from fractional crystallisation.
Aluminium, primary, at plant/RER U	0.8	kg	Primary Al used to dilute scrap to meet purity requirements.
Output			
Refined aluminium	1	kg	

Table 20: Inventory for scenario 3b – fractional crystallisation and distillation as refining method; *Refined aluminium from old scrap, fractional crystallization, with distillation.*

Input	**Amount**	**Unit**	**Comment**
Shredding and sorting, Aluminium	2.04	kg	
Melt aluminium scrap, induction furnace	2/0.98 = 2.04	kg	Assume 2 % loss from melting process.
Secondary casting	1	kg	
Fractional crystallisation, melted old aluminium scrap	1/0.5 = 2	kg	Assume a 50 % yield from fractional crystallisation.
Distillation, 900 C, aluminium melt, old scrap	2	kg	
Output			
Refined aluminium	1	kg	

Table 21: Inventory for scenario 4 – fluxing with Cl_2 gas as refining method; *Refined aluminium from old scrap, fluxing with* Cl_2 *gas.*

Input	**Amount**	**Unit**	**Comment**
Shredding and sorting, Aluminium	1.03	kg	
Melt aluminium scrap, induction furnace	1.005/0.98 = 1.03	kg	Assume 2 % loss from melting process.
Secondary casting	1	kg	
Fluxing with Cl_2 *gas, aluminium melt*	1.005	kg	Assume 0.5 % loss during fluxing.
Output			
Refined aluminium	1	kg	

5. Results

This chapter presents the results obtained during the impact assessment step of the LCA. First the results for the different scenarios will be presented in section 5.1. For each scenario a figure displaying the characterised results is included. These graphs visualise which production step holds the most responsibility for the environmental burden placed upon each impact category. For each scenario the characterised results for the refining step is also included, to trace where the main bulk of impacts stems from. Characterising the results means setting the total impact to be a 100 % and assign the respective share to the production step. For example in Figure 11, which contain the results for scenario 1a – Hoopes process, it can be read that approximately 95 % of the impacts to Global warming potential (GWP) are caused by Hoopes process (the refining step), and about 4 % are caused by the melting step.

In section 5.2., the various sub-processes are investigated to trace the impacts backwards in the system.

In section 5.3. comparisons between scenarios and refining methods have been made. This is also done by characterising the results. When using characterisation to compare results, the process or scenario with the highest impact to a category is set to be 100 % and then the impacts from the other processes or scenarios is measured according to this. In addition, as according to the goal of this assessment, the refining scenarios are compared to primary aluminium production and a general secondary production route. The processes used to these comparisons are existing ecoinvent processes. This comparison will demonstrate how the introduction of a refining step on a secondary production route will affect the environmental impacts.

The fourth section of this chapter aims to identify which important impact categories are most important related to the context of this thesis. This is investigated using ReCiPe Midpoint and ReCipE Endpoint indicators. For both investigations is from a hierarchical perspective using average European weighting and normalisation.

The last section provides an investigation related to the validity of these results by comparing them to the existing processes related to this topic in ecoinvent, and by discussing some of the uncertainties related to the data.

Additional results are found in Appendix C.

5.1. Scenario results

This section presents the characterised results for each scenario. These results will show which of the four main production steps in Figure 12 contributes to the main bulk of environmental impacts.

Scenario 1a – Hoopes process as refining method

Figure 13 displays the results for scenario 1a. Figure 13 includes all the impact categories covered by the method used. It is evident that the electrolysis is the main contributor to all impact categories. For nearly all impact categories approximately 90 % of all impacts are caused by Hoopes process alone. The exceptions are water resource depletion, the cumulated use of renewable energy from wind, solar and geothermal sources and the cumulated use of non-renewable biomass. For these three impact categories the casting process contributes more than it does for the remaining categories. The second largest contributor to most impact categories is the melting process, which accounts for about 4 % of the impacts. It is clear that the sorting and shredding does not hold much responsibility for the environmental impacts caused by this scenario. From Table A in Appendix C, one can find that the contribution from this process is approximately 0.5 % to all impact categories.

The most important result read from Figure 13 is that it is clearly the refining step that is most responsible for the environmental impacts caused by this scenario. Based on this the characterised results for the refining step alone is included in Figure 14. Hoopes process is modelled including the use of electricity, infrastructure and disposal of dross from the electrolysis. Figure 14 clearly shows that it is the use of electricity that is accountable for the main bulk of impacts. The dross disposal is insignificant for this process; it is less than 0.001% for all categories. An interesting result is the seemingly positive effect from infrastructure on land use. Investigating of the infrastructure process reveals that this happens due to assumptions related to re-cultivation of land after demolition of the plant. Land use is measured in kg C deficit, which means avoided carbon. If it is assumed that the plant is built on uncultivated land, no carbon absorbing biomass, and that this land is cultivated after demolition, the total effect on land use would be positive.

The numerical results for Hoops process are included in Table B in Appendix C.

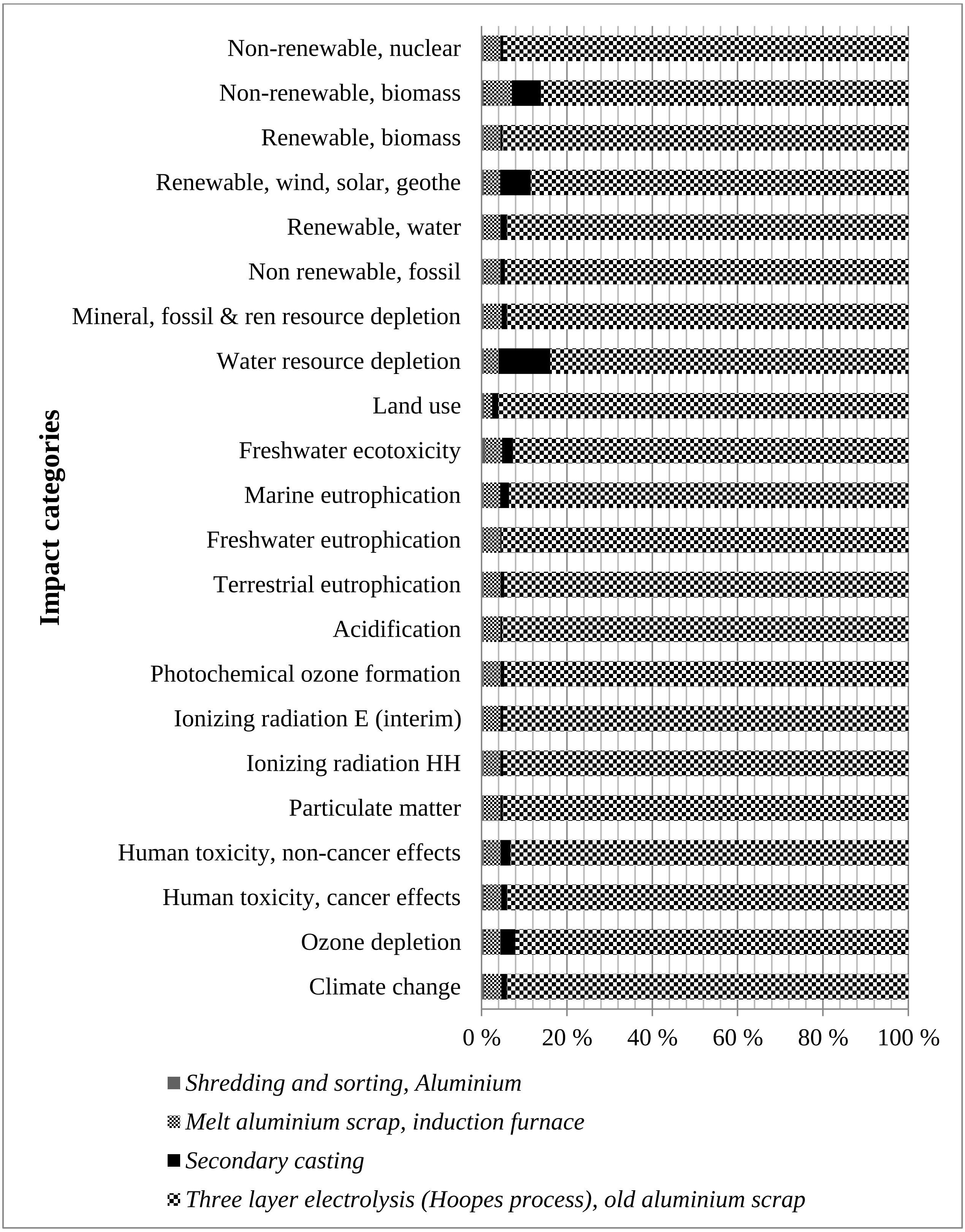

Figure 13: Characterised results for scenario 1a – Hoopes process as refining method. 100 % is the total environmental burden placed upon each impact category by the respective production scenario.

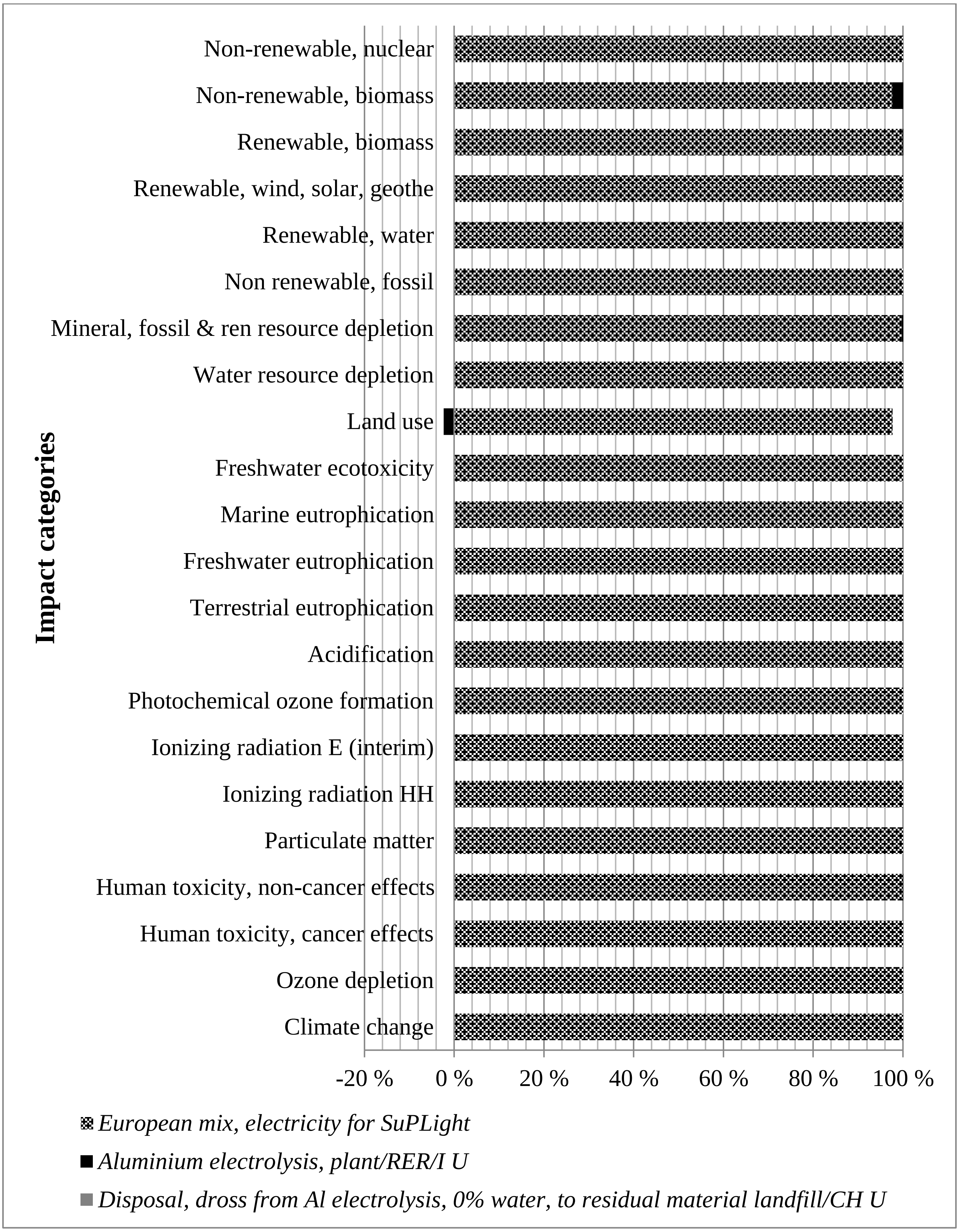

Figure 14: Characterised results for Hoopes process. 100 % is the total environmental burden placed upon each impact category by the respective refining method.

Scenario 1b – Adjusted three-layered electrolysis

The characterised results for scenario 1b are given in Figure 15. It is stated in the scenario descriptions that this scenario is not very different from the previous scenario. The energy-need is adjusted to fit use on scrap metal and meet lower purity demands. Therefore it is not surprising that the results resemble the results for the previous scenario. The difference between the scenarios is the share of impacts caused by the refining method. In this scenario the refining method is responsible for 80-90 % of the impacts to all categories except for water resource depletion, the cumulated use of renewable energy from wind, solar and geothermal sources and the cumulated use of non-renewable biomass. For these the categories the casting process holds a larger share of the responsibility than it does for the remaining categories. In this scenario the melting step accounts for approximately 8 % of the impacts to all categories. The sorting and shredding accounts for 0.9 – 2 %, and is still quite insignificant.

Since it is clear for this scenario that the refining step is the main contributor to all categories, characterised results for the adjusted three-layered electrolysis is included in Figure 16. Numerical results for this refining method are included in Table D in Appendix C. This refining method is modelled similar to Hoopes process and therefore the results resembles the results obtained for Hoopes process; namely that the electricity use is the main contributor to all impact categories. The effect on land use from the infrastructure sticks out more, but by comparing Table B and Table D in Appendix C, it is evident that the actual impacts from the infrastructure are the same. The impacts from dross disposal are still negligible, and are also the same as for the previous scenario.

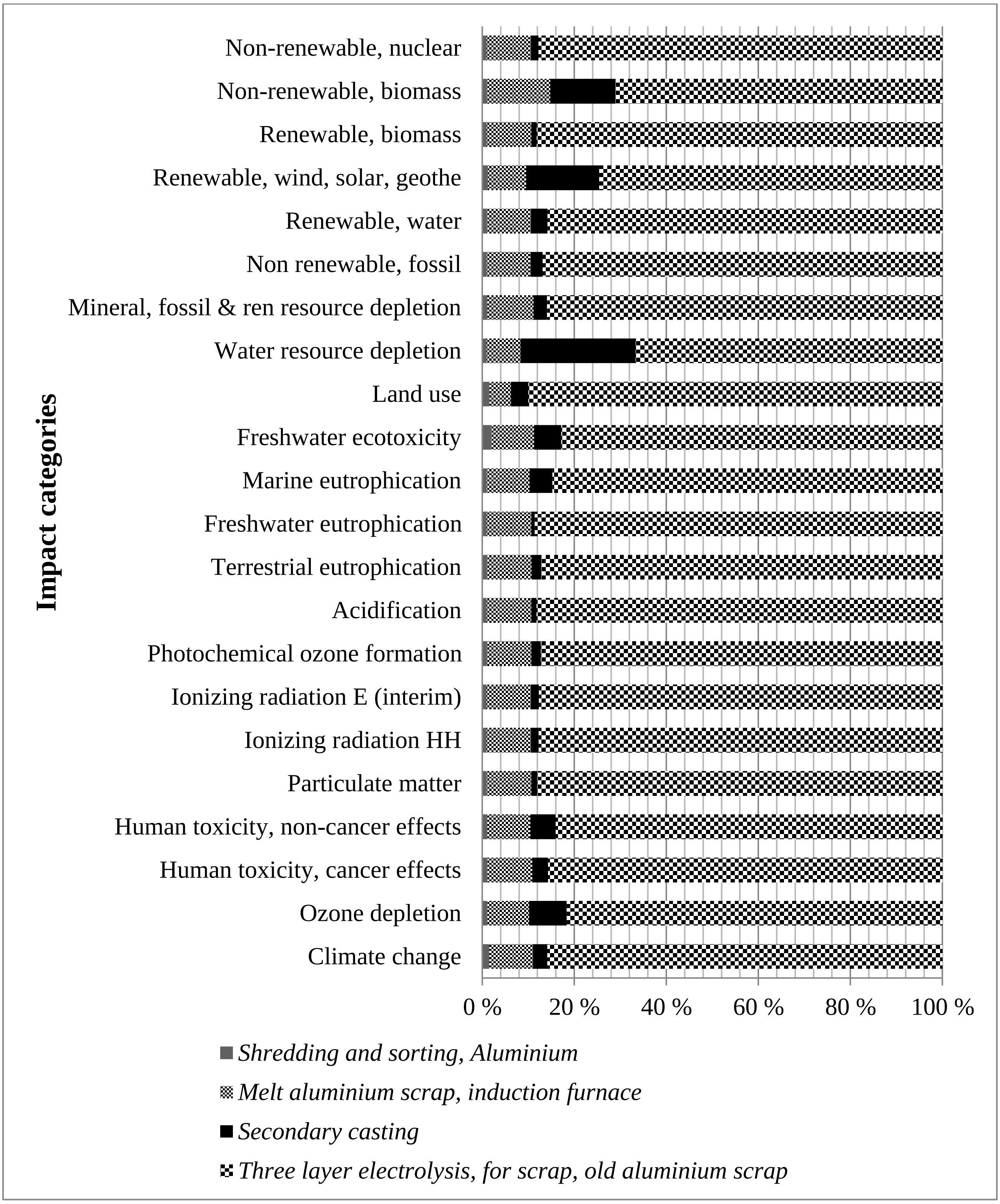

Figure 15: Characterised results for scenario 1b – adjusted three-layer electrolysis as refining method. 100 % is the total environmental burden placed upon each impact category by the respective production scenario.

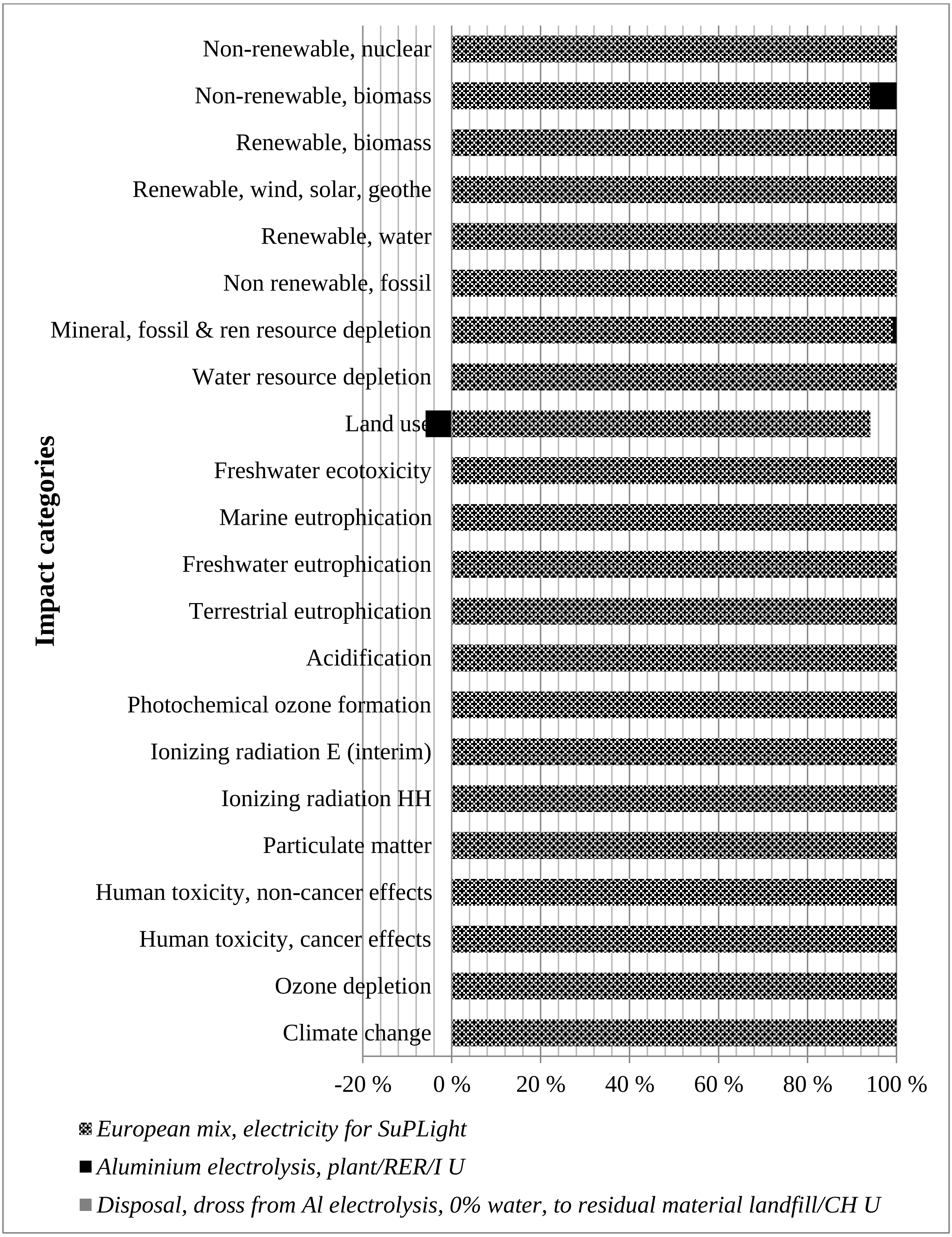

Figure 16: Characterised results for the adjusted three-layered electrolysis. 100 % is the total environmental burden placed upon each impact category by the respective refining method.

Scenario 2- Low temperature electrolysis

The low temperature electrolysis has an even lower energy use than the adjusted three-layered electrolysis. The fact that the melting process is omitted from this scenario reduces the energy use in this scenario even further. The result for this scenario is seen in Figure 17, and it is evident that the refining process is the main contributor to all impact categories for this scenario as well. The casting process takes on a larger share of the responsibility of impacts, but the numerical value is the same. Numerical results for this scenario are included in Table E in Appendix C.

Characterised results for the refining method used in this scenario are displayed in Figure 18. Again it is evident that the electricity use is the main contributor to all impact categories. A larger effect from the infrastructure is spotted, but again the numerical results are the same as for the two previous scenarios. The impacts from dross disposal are still negligible, and are also the same as for the two previous scenarios.

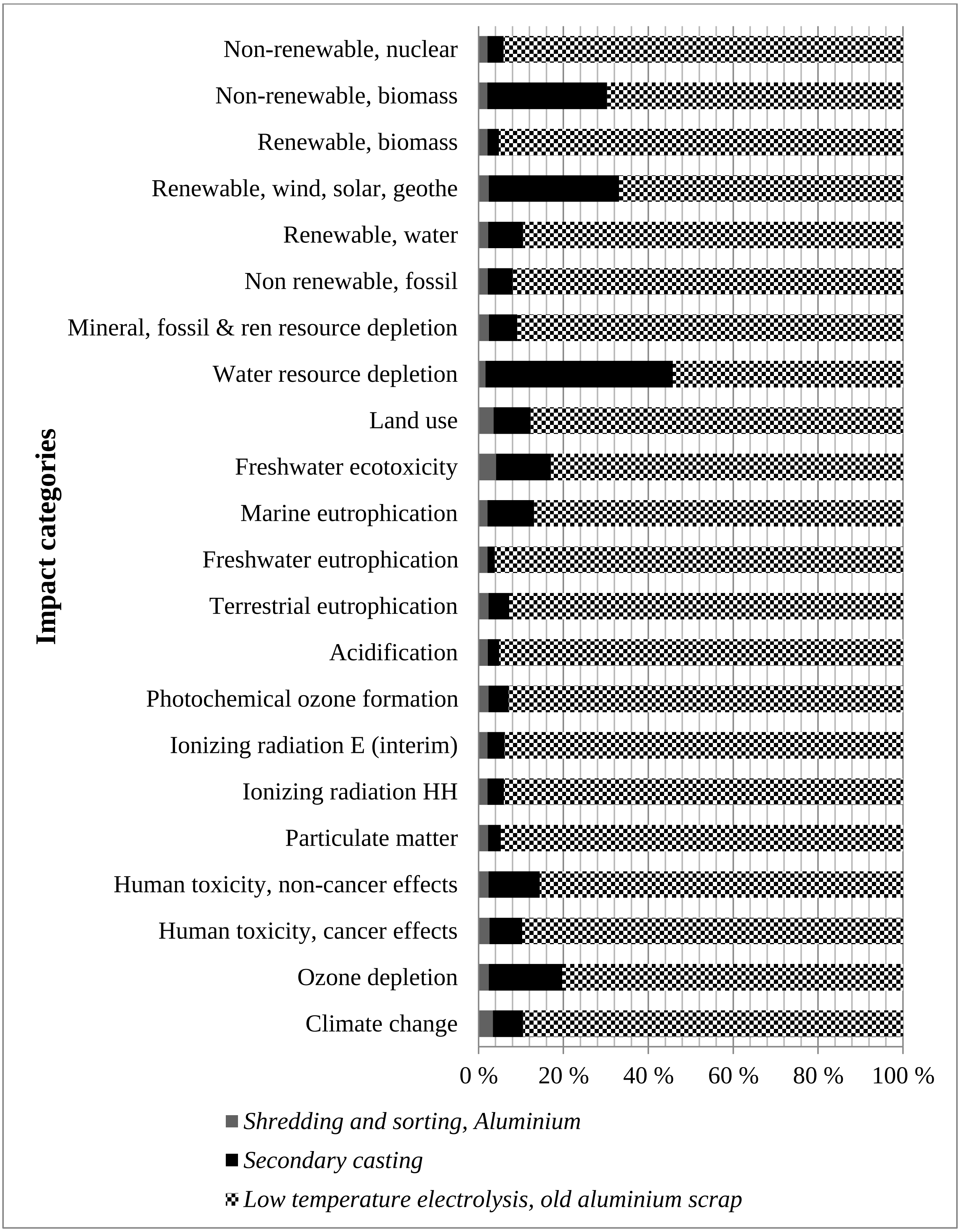

Figure 17: Characterised results for scenario 2 – low temperature electrolysis as refining method. 100 % is the total environmental burden placed upon each impact category by the respective production scenario.

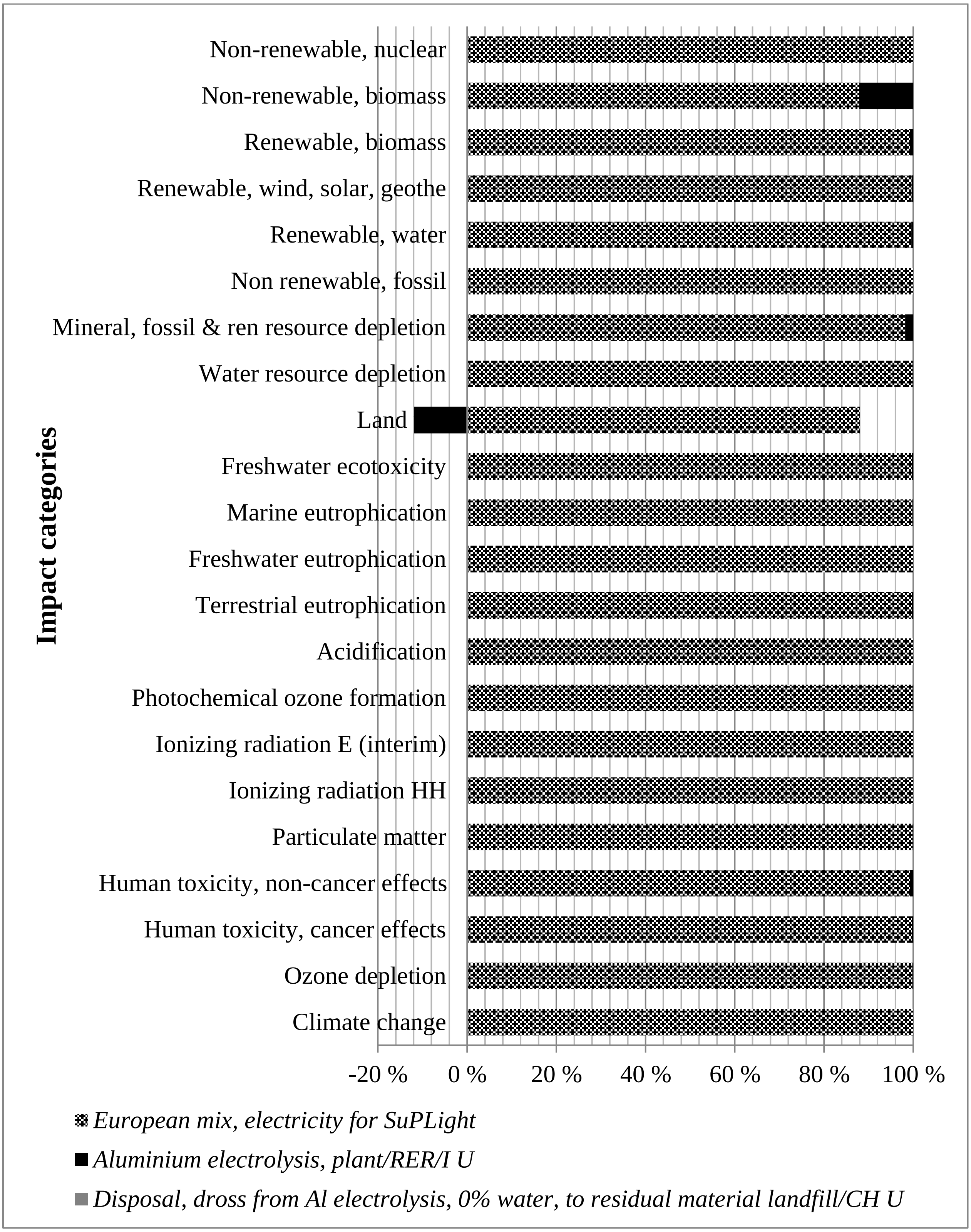

Figure 18: Characterised results for low temperature electrolysis. 100 % is the total environmental burden placed upon each impact category by the respective refining method.

Scenario 3a - Fractional crystallisation and dilution as refining method

Characterised results for this scenario are included in Figure 19. For this scenario fractional crystallisation alone was not able to meet the purity requirements set in Table 2, therefore some dilution was required. The partitioning between refined scrap and primary aluminium is 20 and 80 % respectively. From Figure 19 it is clear that the primary aluminium is responsible for the main impacts caused by this scenario. Another noticeable result for this scenario is that the fractional crystallisation has lower contributions to the total impacts than the melting process. The shredding and sorting process is still negligible and accounts for less than 1 % for all impact categories.

The process for primary metal used to dilute the scrap is an existing ecoinvent process. It is based on average technology for Europe and is modelled based on LCI work done by EEA, as described in chapter 3. Even through this process is responsible for the largest share of impacts in this scenario, fractional crystallisation is the refining process studied further. This is mainly because the overall goal of this assessment is to evaluate possible refining processes and because impacts from primary production are fairly known and mentioned in the introduction of this thesis. In relation to primary production the production of red-mud and high energy use are the main factors.

The characterised results for fractional crystallisation are displayed in Figure 20. This process is modelled including electricity use and infrastructure. It is evident that it is divided which of the two processes contributes the most to the various impact categories. It is noticeable that the same positive effect on land use from infrastructure occurs for this scenario as well, even though the infrastructure is modelled using a different process than for previous scenarios. If the impacts from the two different infrastructures are compared it is found that the impacts from the *Metal working factory* are larger than the impacts from the *Aluminium electrolysis, plant*. Numerical values for these processes are found in Table H and F respectively.

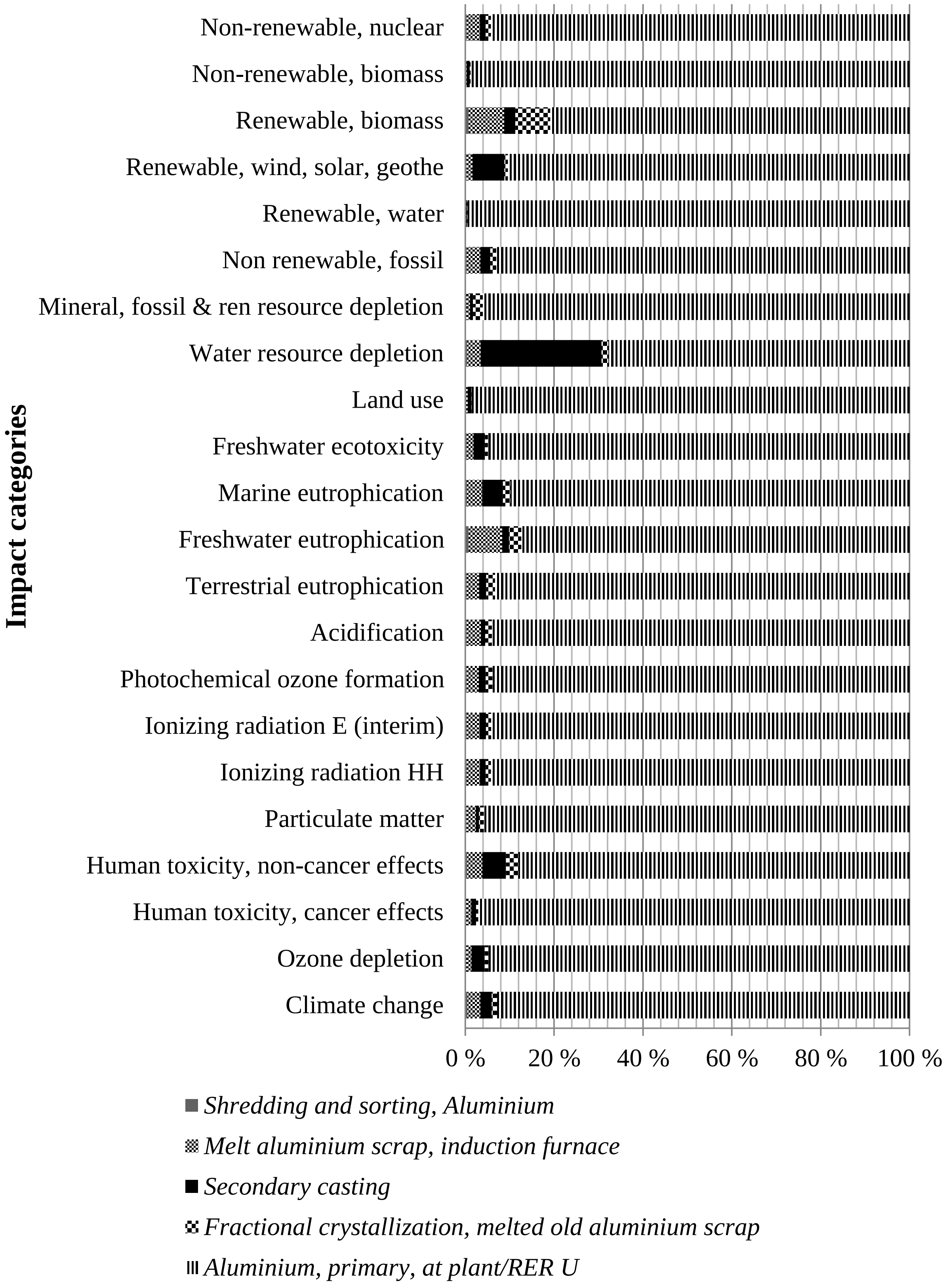

Figure 19: Characterised results for scenario 3a – fractional crystallisation and dilution as refining method. 100 % is the total environmental burden placed upon each impact category by the respective production scenario.

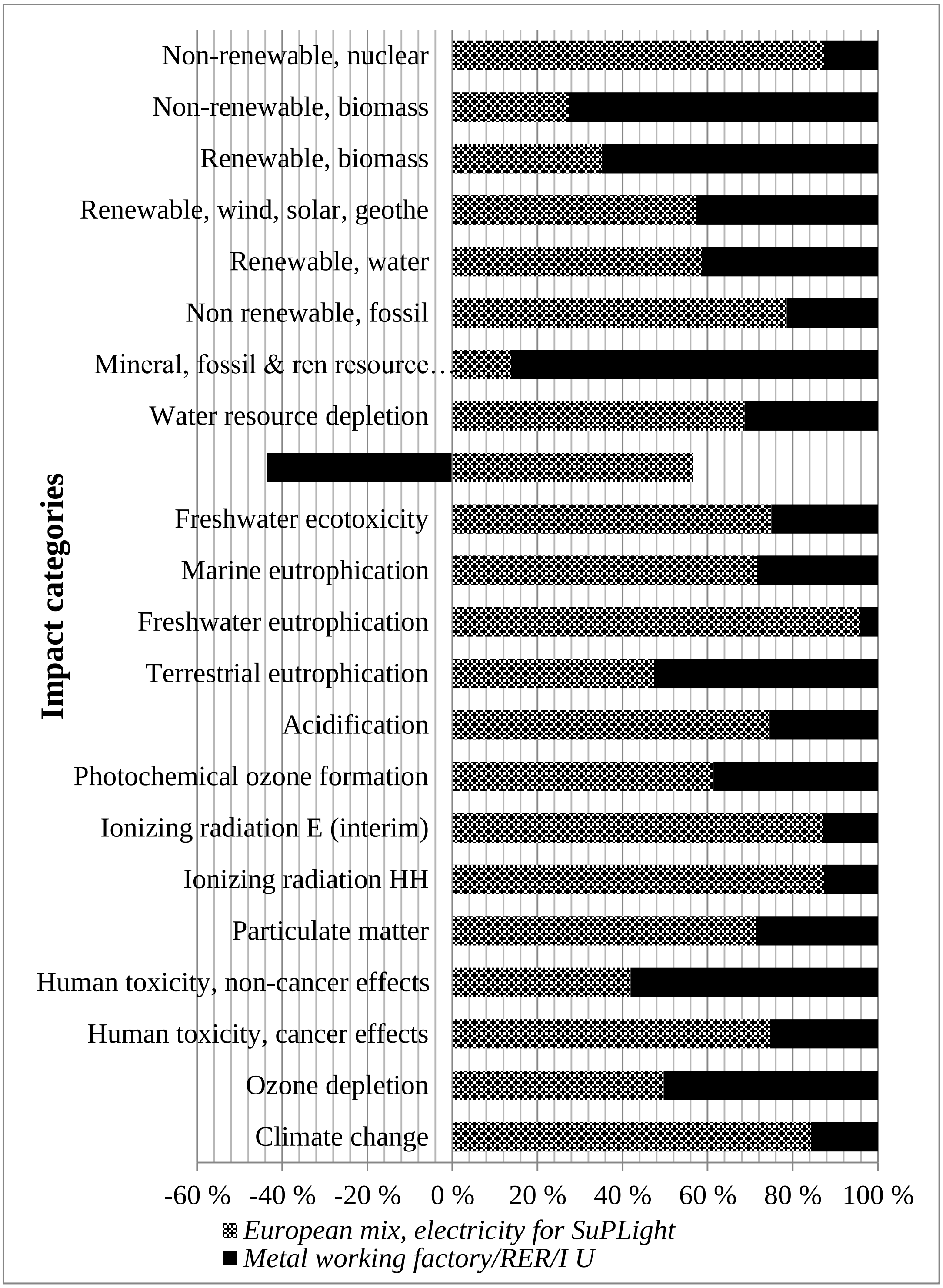

Figure 20: Characterised results for fractional crystallisation. 100 % is the total environmental burden placed upon each impact category by the respective refining method.

Scenario 3b - Fractional crystallisation and distillation as refining method

Characterised results for this scenario are included in Figure 21. This scenario combines distillation with fractional crystallisation to obtain the given purity demands given in Table 2. From Figure 21 it is evident that the main bulk of impacts stems from the distillation. And since the fractional crystallisation was investigated for the previous scenario, a closer look at distillation is included here. Characterised results for distillation are included in Figure 22. It is evident that the electricity accounts for the largest share of impacts. For this scenario the melting process accounts for a larger share of the impacts than fractional crystallisation, which is similar to the previous scenario. For this scenario the impacts from melting is approximately 20 % for most impact categories and around 4 % from the sorting and shredding process.

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An important difference between scenario 3a and scenario 3b is that since dilution is applied in scenario 3a only 20 % scrap is utilised. This effect the amount of impacts related to the scrap. The impacts from shredding and sorting, melting and refining of the scrap will therefore be much lower for scenario 3a than for scenario 3b.

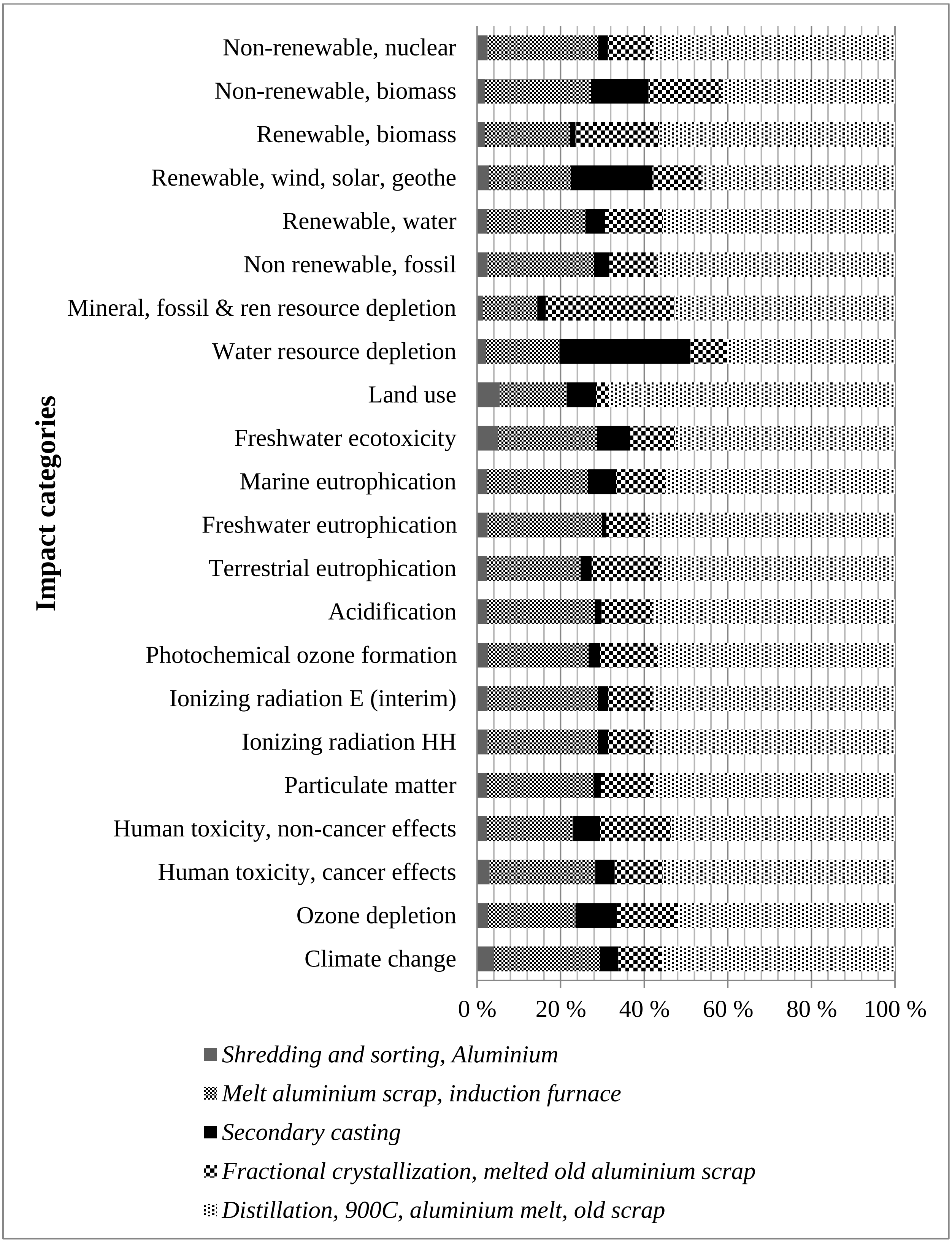


Figure 21: Characterised results for scenario 3b – Fractional crystallisation and distillation as refining method. 100 % is the total environmental burden placed upon each impact category by the respective production scenario.

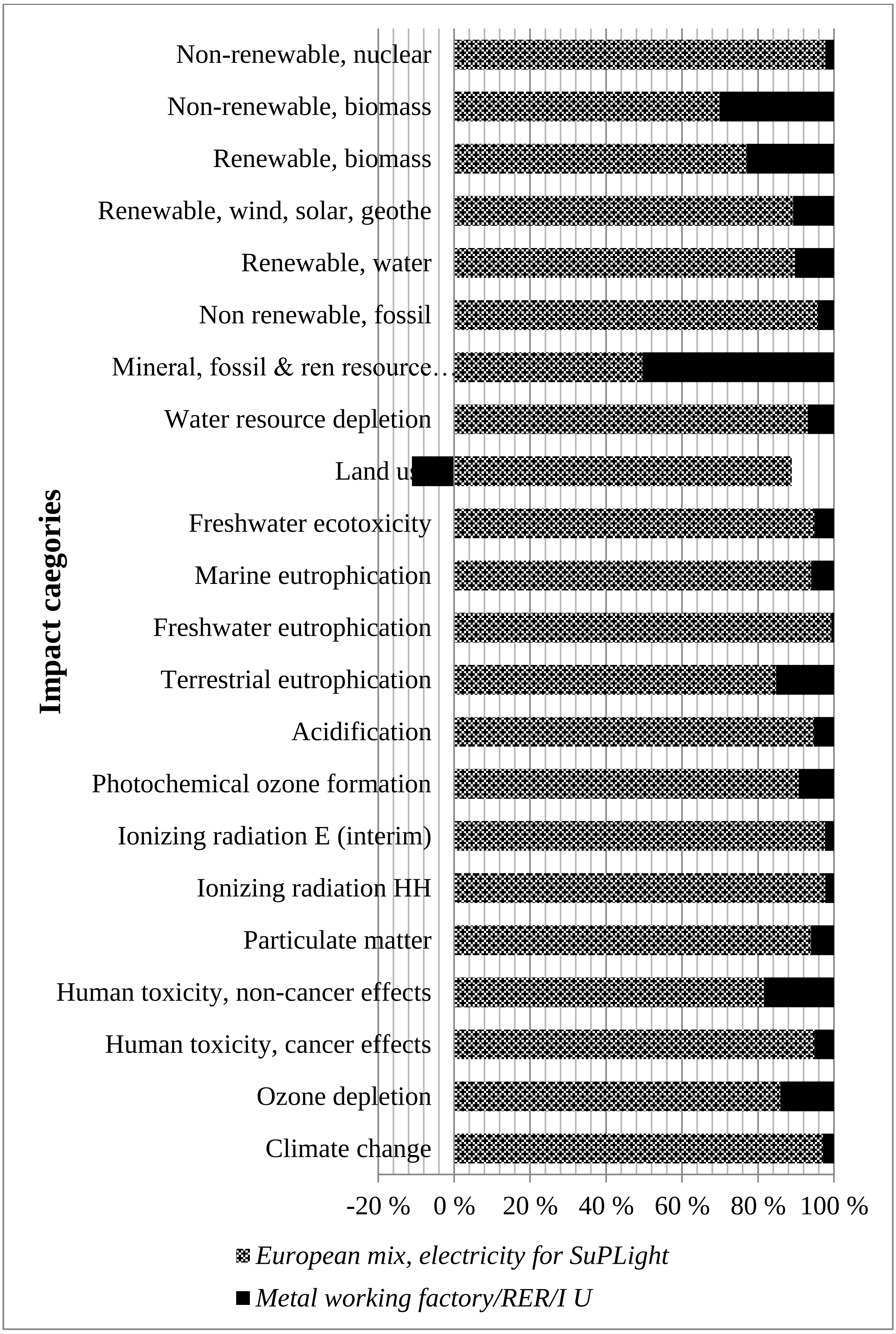


Figure 22: Characterised results for distillation. 100 % is the total environmental burden placed upon each impact category by the respective refining method.

Scenario 4- Fluxing with Cl_2 gas as refining method

As mentioned in the description of this scenario, this cannot be understood as a complete scenario since the purity demands are not met by this refining method. However it is perceived interesting to include result a scenario which utilise fluxing with Cl_2 gas since this a widely used technique used in industry. Since the total impacts for this scenario is lower than for the other scenarios the sorting and shredding, melting and casting processes are responsible for a larger share of the total impacts. The characterised results for this scenario are displayed in Figure 23. Fluxing with Cl_2 gas as a refining method is also investigated further and characterised results for this process alone is included in Figure 24. From this figure it is evident that for most impact categories the electricity is most responsible for the total impacts. Since chlorine is used in this process a process to model disposal of hazardous waste is included. This process is only responsible for less than 0.001 % of the impacts for all impact categories. It is noticeable that the chlorine used as a flux accounts for a large share of the impacts to two of the categories; namely ozone depletion and the cumulated use of renewable wind, solar and geothermal energy. When traced backwards this is related to the energy mix used in chlorine production and some release of Cl which is damaging for the ozone layer.

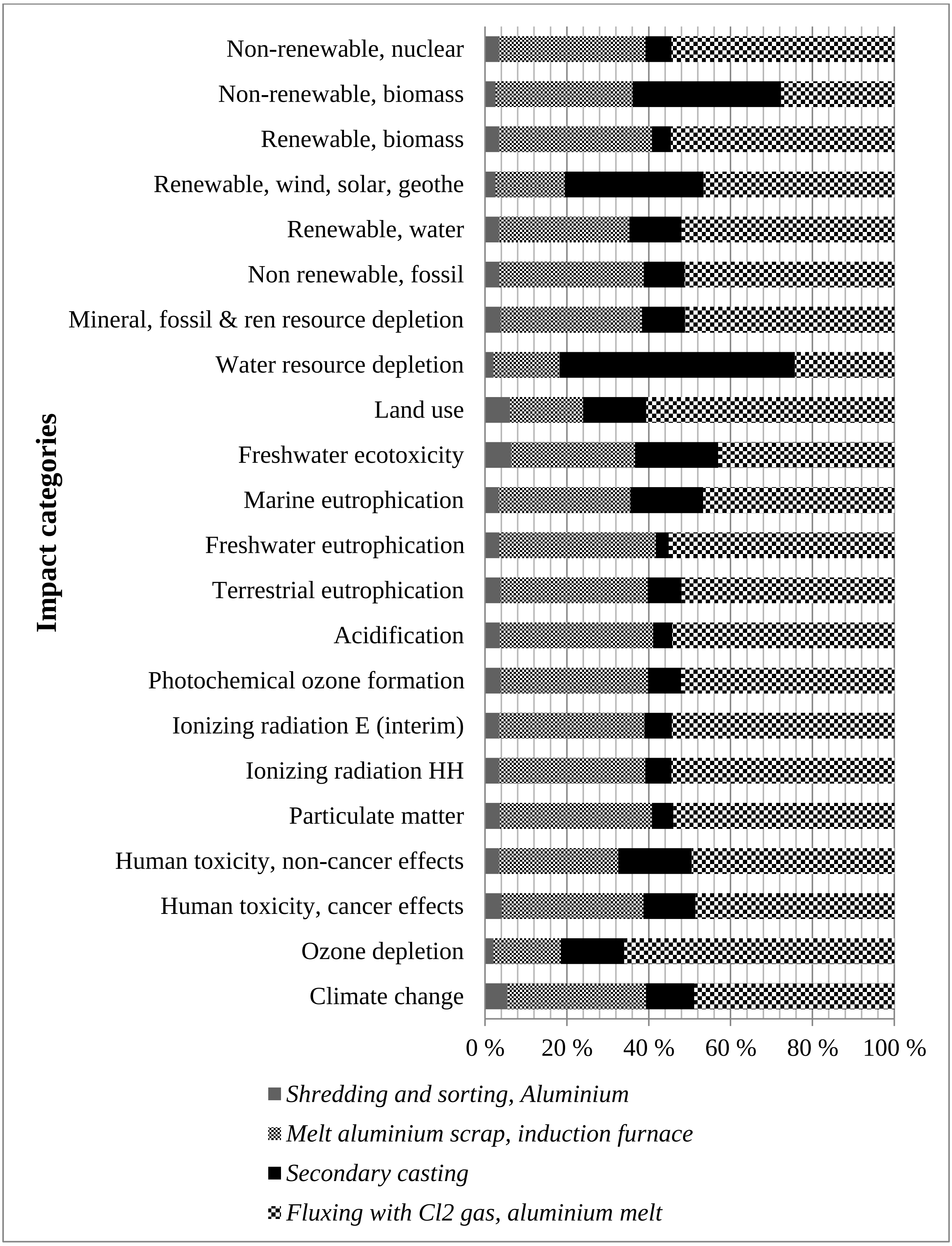


Figure 23: Characterised results for scenario 4 – fluxing with Cl_2 gas as refining method. 100 % is the total environmental burden placed upon each impact category by the respective production scenario.

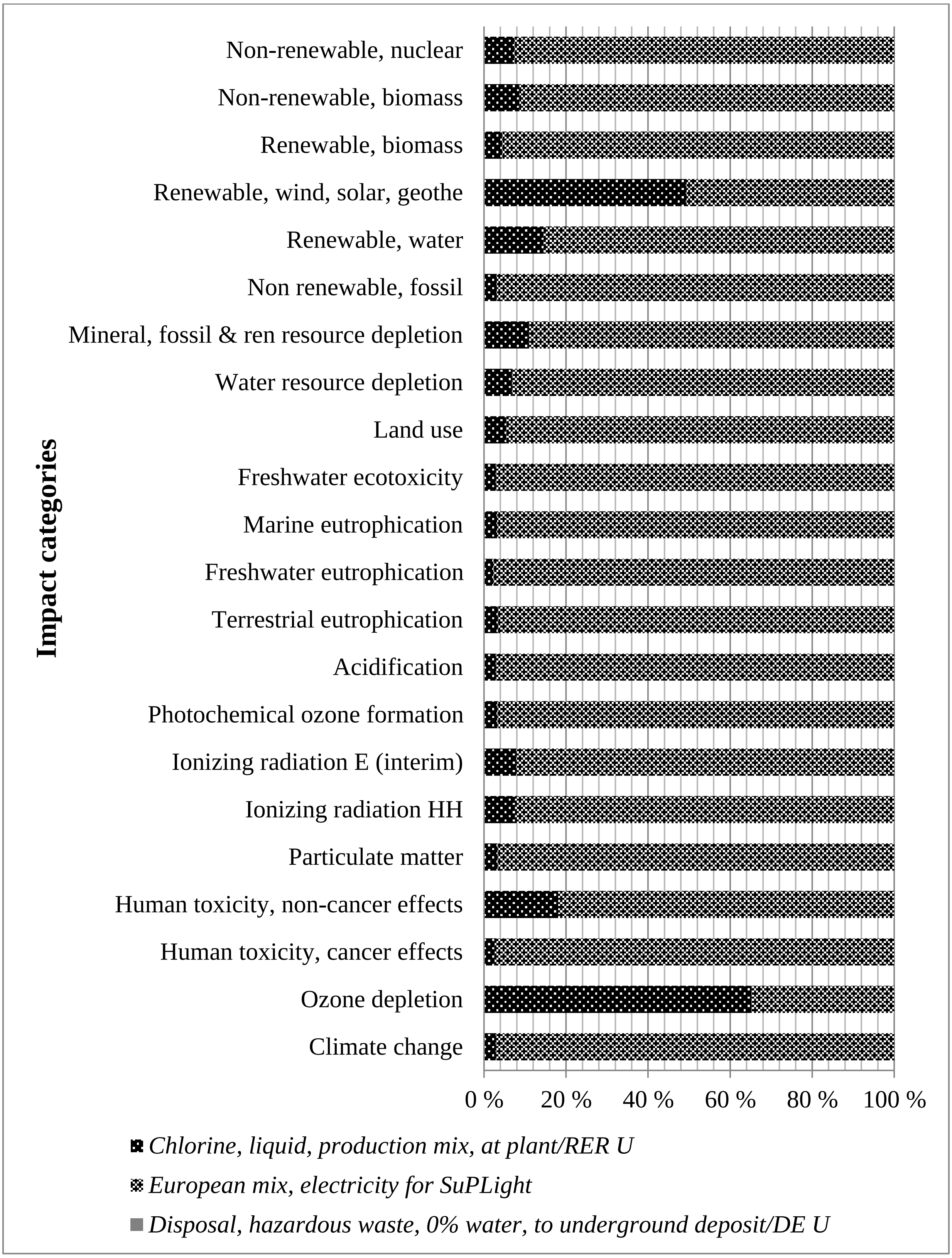


Figure 24: Characterised results for fluxing with Cl_2 gas. 100 % is the total environmental burden placed upon each impact category by the respective refining method.

5.2 Results for individual processes

Collection, sorting and shredding

In the previous section it was found that the sorting and shredding process only accounts for a very small part of the impacts to all impact categories for all scenarios. Therefore in depth results for this process are not included. Some backward tracing of impacts do however reveal that in this process the shredding accounts for most of the impacts and that the electricity used during shredding is the main contributor. The two main disposal processes are disposal of plastics and rubber. Shredding of ELV scrap, disposal of rubber and disposal of plastics account for between 34 % and 70 % of the impacts for all impact categories.

Melting process

Throughout the execution of analysing the result for the three cases, the melting process without a salt layer is used. This is due to two arguments; first it is stated that a salt layer is mainly used when producing high purity aluminium, which is not the case here, and secondly there are large uncertainties related to the salt-layer which can result in results that are more wrong than necessary. Characterised results for the melting process used during the impact assessment are included in Figure 25. It is clear that the electricity is responsible for the largest share of the impacts associated with the melting process. There are also some positive effects on land use from the infrastructure, due to reasons discussed earlier. It is also evident that the waste disposals accounted for does not cause much environmental impact.

Casting process

The only inputs to this process are infrastructure and operation of the metal factory. Characterised results for this process are included in Figure 26. It is evident that it is the operation of the plant that is the dominating contributor to all impact categories. Backward tracing of the operation process reveal that the main impacts occur from energy use to for example lighting and tap water use.

Electricity production mix

As it is evident for all refining processes and most sub-processes that it is electricity use that causes the most impacts, it is interesting to investigate the electricity production mix used during the assessment. It is stated in chapter 4 that the mix is composed of roughly 28 % Czech production, 11 % Hungarian production, 51 % Polish production and 10 % Slovak production. Tracing the impacts from these four productions reveals that they are mainly due to power production based on fossil fuels like natural gas, coal and lignite.

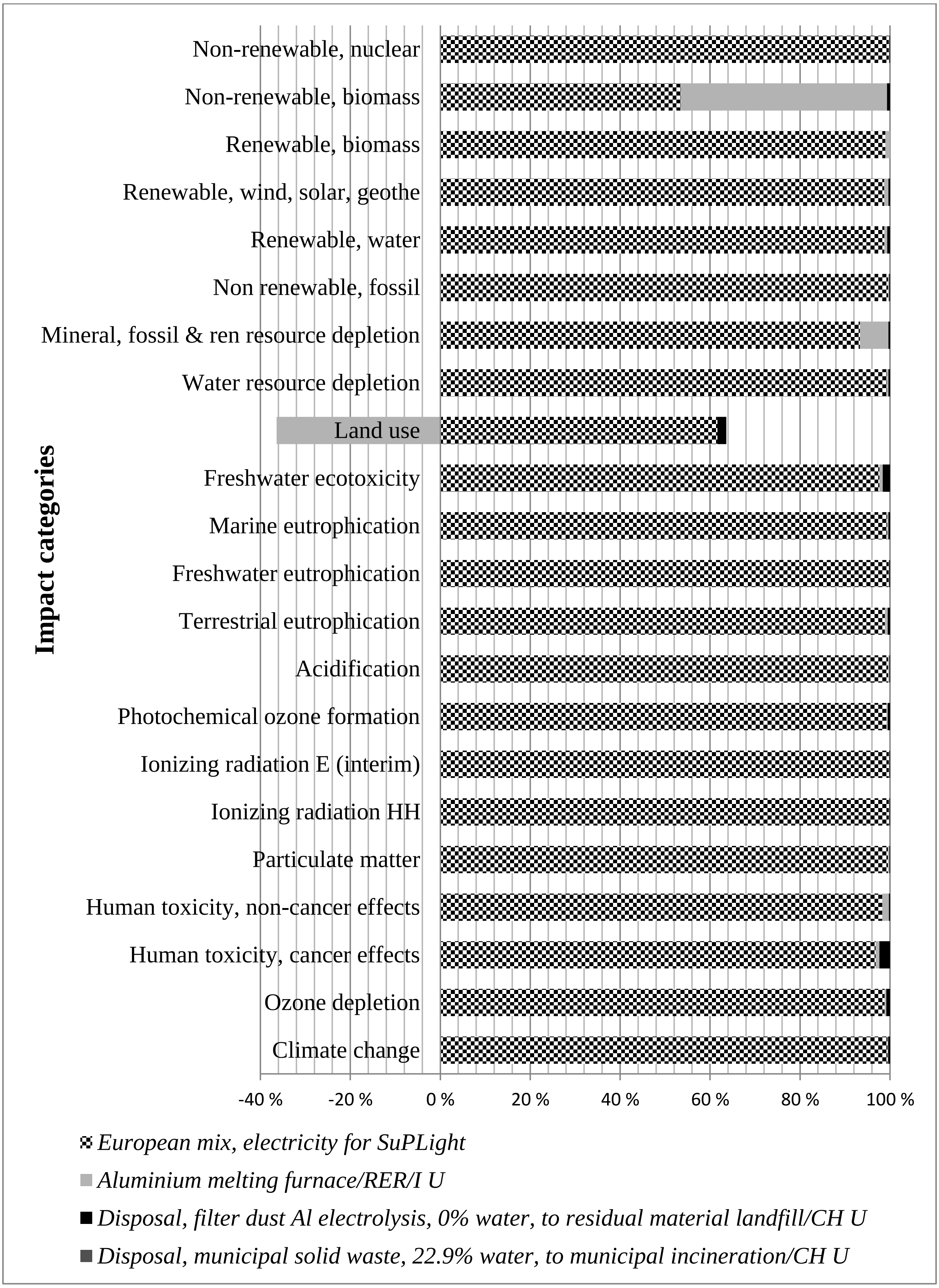


Figure 25: Characterised results for melting process. 100 % is the total environmental burden placed upon each impact category by the melting process.

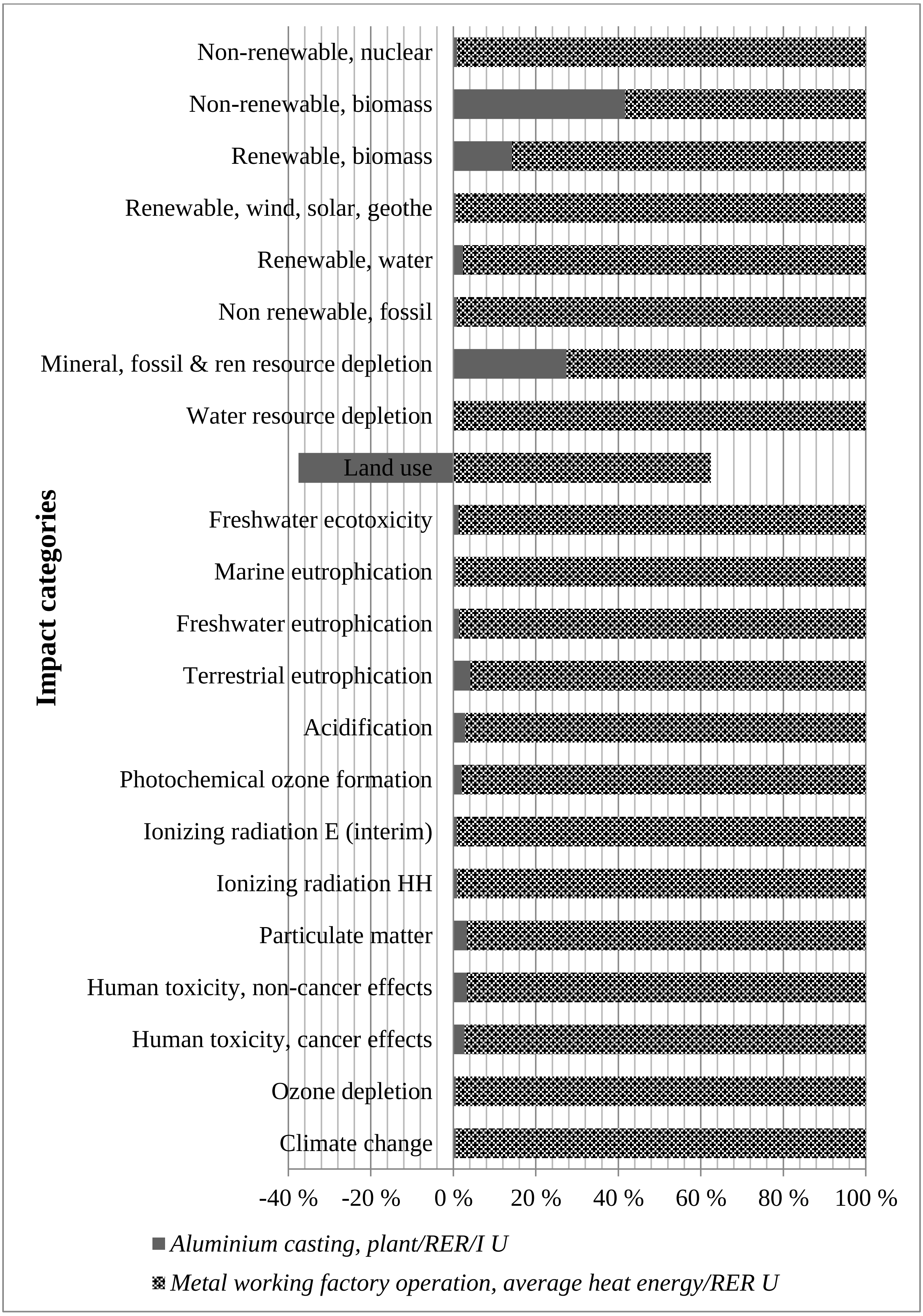


Figure 26: Characterised results for casting process. 100 % is the total environmental burden placed upon each impact category by the casting process.

5.3 Comparisons

A goal of this assessment is to be able to compare different refining options with each other and primary production.

Comparing the six production scenarios

Figure 27 clearly shows that scenario 1a, which utilises Hoopes process as the refining method, has the high environmental impact compared to the other scenarios. This is predicted though, due to the very high energy use compared to the other production scenarios and primary aluminium production. The scenario generally having the second highest impacts is scenario 3a, which utilises fractional crystallisation and dilution. This is not surprising since the finished alloy is composed of 20 % scrap and 80 % primary metal, and the high energy use during primary production is known. After scenario 3a scenario 1b has the highest impacts. This is the scenario utilising the adjusted three-layer electrolysis. Even though this scenario has lowered the energy needed during electrolysis a lot compared to Hoopes process, the energy used is still high compared to the other scenarios. The scenarios using low temperature electrolysis, fluxing and fractional crystallisation with distillation are the scenarios generally having the lowest impacts to all categories.

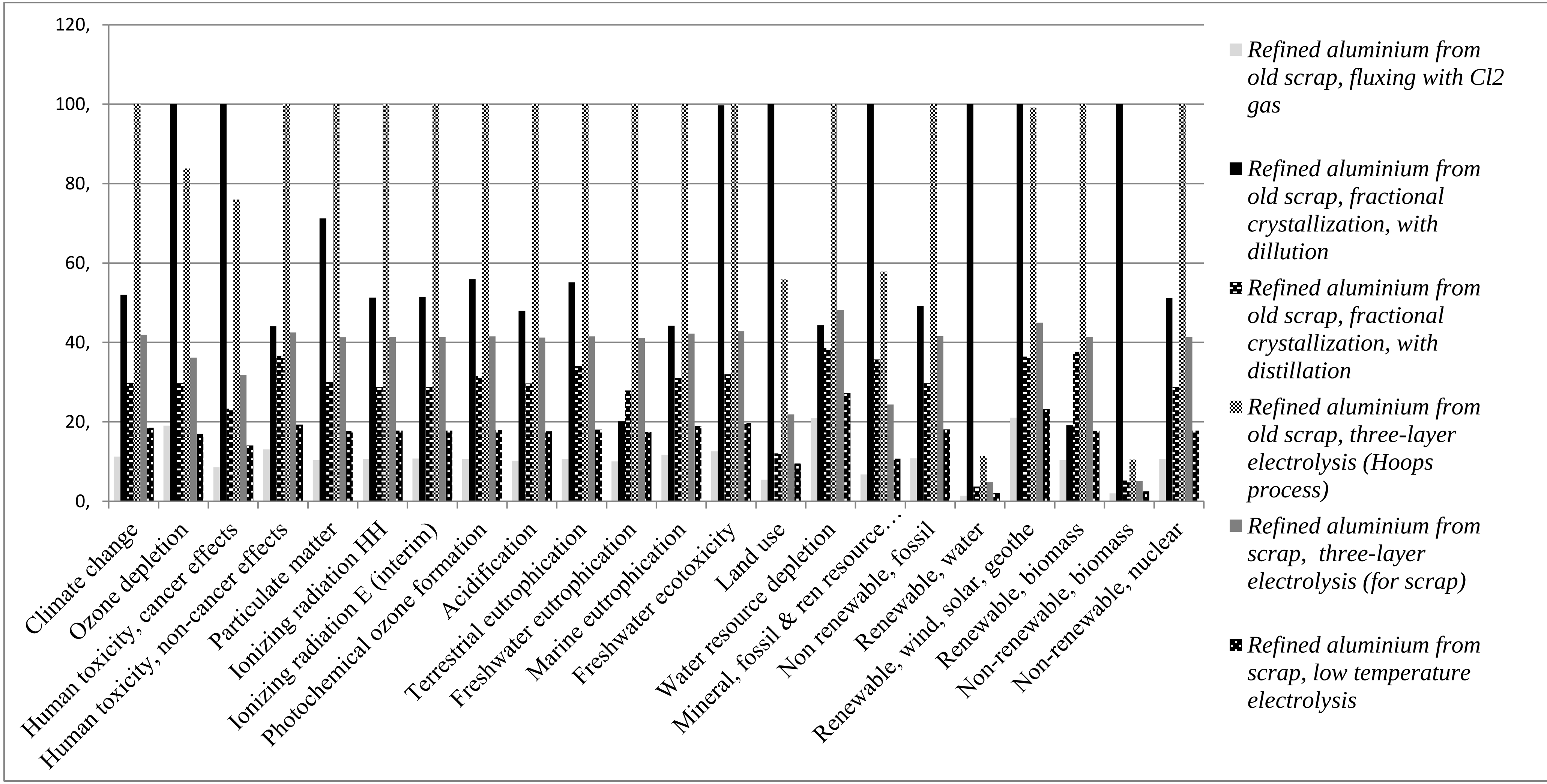


Figure 27: Comparing the six different production scenarios for post-consumer scrap which includes a refining step. 100% represent the highest impact to an impact category caused by one of the respective scenarios.

Comparison between primary production and refining of scrap

A goal of this assessment is to compare the production of pure aluminium utilising post-consumer scrap with primary production and traditional secondary production. This is done in two steps. First the scenarios including electrolysis are compared to both primary and secondary production. This is included in Figure 28. This comparison shows that scenario 1a have the highest impacts to most categories. Primary aluminium has the highest impacts to cumulated use of non-renewable biomass, renewable wind, solar and geothermal sources, renewable water sources, land use, freshwater ecotoxicity, ozone depletion and human toxicity (cancer effects). A surprising result from this comparison is that the traditional secondary production has the highest impacts to mineral depletion and human toxicity (non-cancer effects). Scenario 1b, which uses the adjusted three-layer electrolysis, has lower impacts overall compared to both primary production and scenario 1a. Scenario 2, using low temperature electrolysis, has even lower impacts. This trend is not surprising since it was found in the previous section that energy use is very closely linked to the environmental impacts associated with the various scenarios.

Figure 29 contains the comparison between production scenarios which includes fractional crystallisation with primary production and traditional production route for secondary aluminium. This comparison confirms the high impact caused by secondary production to human toxicity (non-cancer effects) and mineral depletion. Based on this surprising result, the process used to model the traditional production route for secondary scrap was studied more in-depth. It is found that this process includes alloying with Zn and Cu. The process used to model primary production does not include any alloying with other metals. The production scenarios developed for this thesis does not include any alloying either. The alloying of Zn in the secondary production process causes the largest impacts to human toxicity (non-cancer effects) and mineral depletion. Therefore the difference in inclusion of alloying with metals is found as the reason for the surprising results.

Figure 29 also show that scenario 3a, which utilises dilution together with fractional crystallisation, and primary production has very similar impacts. This is not surprising since the dilution is 80 %. For three impact categories scenario 3b has larger impacts than scenario 3a. These categories are cumulated use of renewable biomass, water resource depletion and freshwater eutrophication. This is probably due to the fact that infrastructure is included for both fractional crystallisation and distillation.

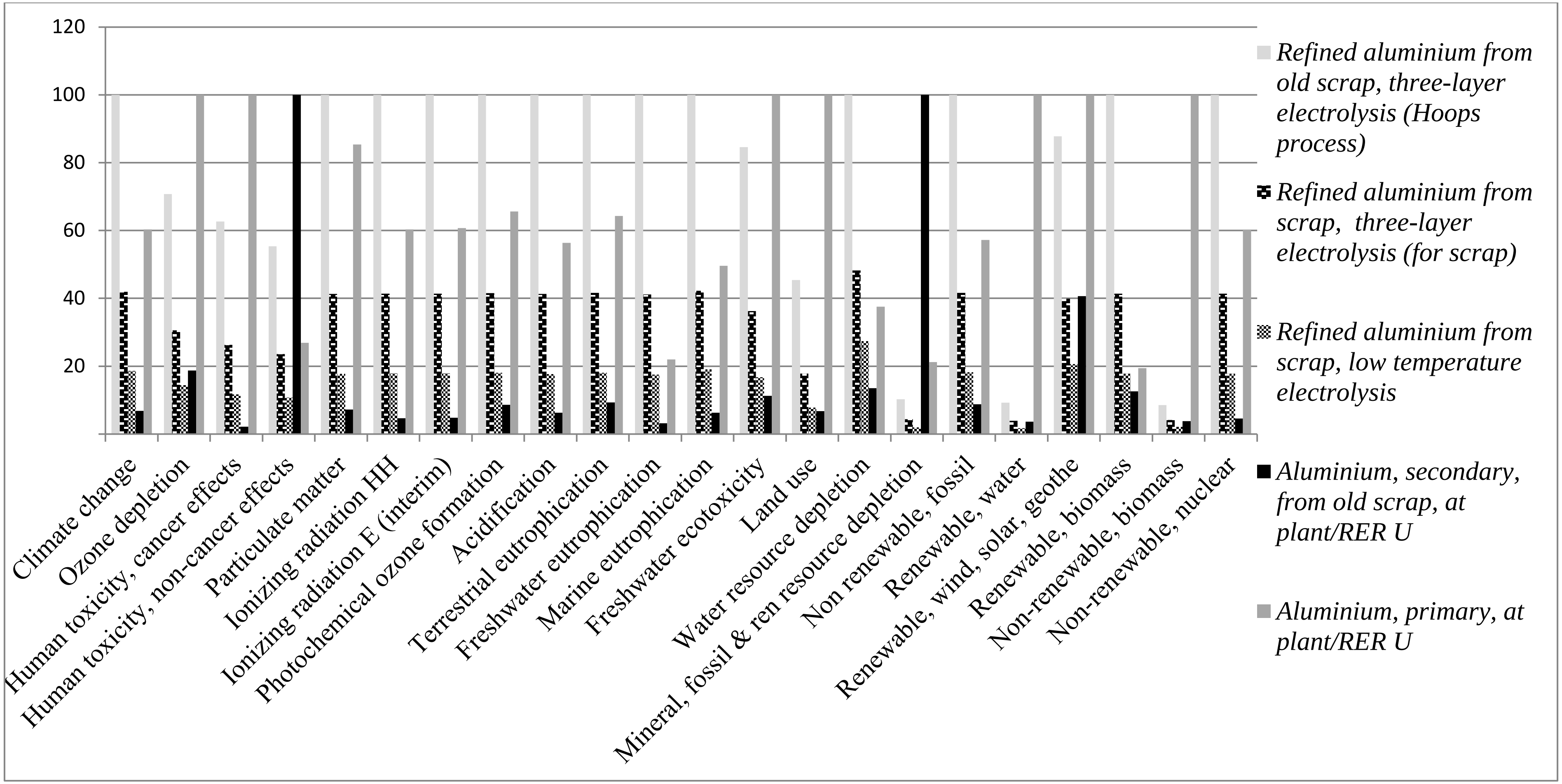


Figure 28: Comparing production scenarios, which include electrolysis, with primary production and traditional secondary production. 100 % represents the highest impact caused by the respective production route.

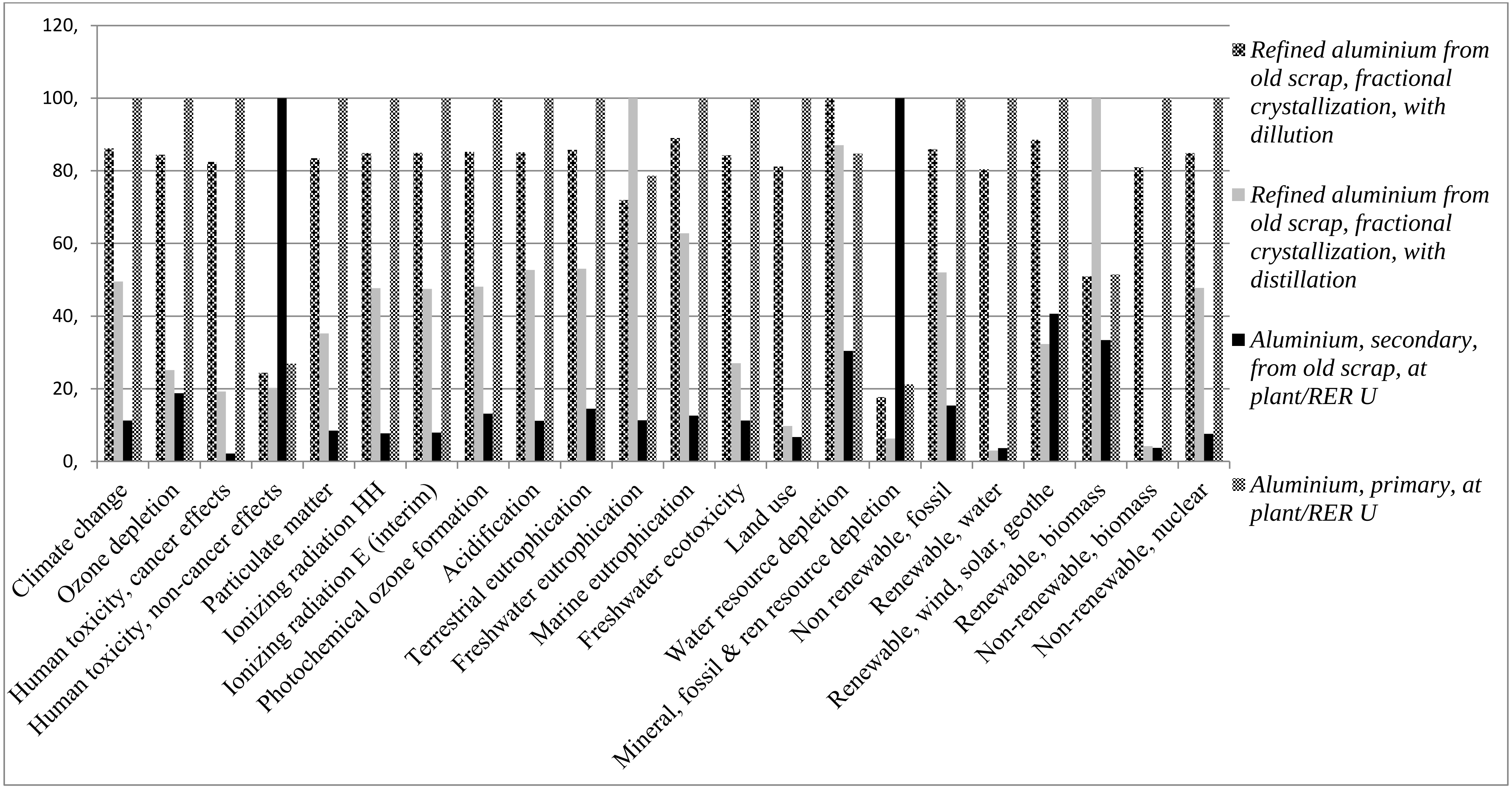


Figure 29: Comparing production scenarios, which include fractional crystallisation, with primary production and traditional secondary production. 100 % represents the highest impact caused by the respective production route.

5.4 Important impact categories

An interesting question which has not been addressed yet is which impact category is more important to consider when making a conclusion based on the results presented so far. To establish this, the ReCiPe method is used. This method converts the long list of results into some indicators which capture the severity to each impact category based on a set of weightings.

One analysis is based on endpoint indicators and one is based on midpoint indicators. Endpoint indicators tend to be more uncertain and midpoint indicators tend to have a shorter perspective. For both a hierarchical perspective is used together with average weighting and normalisation according to European standards. These results are included in Figure 30 and 31.

Figure 30, which is based on midpoint indicators, indicate that freshwater eutrophication, human toxicity, freshwater and marine ecotoxicity and natural land transformation are the most affected indicators.

Figure 31, which is based on endpoint indicators, indicate that global warming, human toxicity, particulate formation and fossil depletion are the most affected indicators.

Based on these two figures is can be suggested that impacts to global warming potential, human toxicity, ecotoxicity and energy use from fossil sources should prioritised.

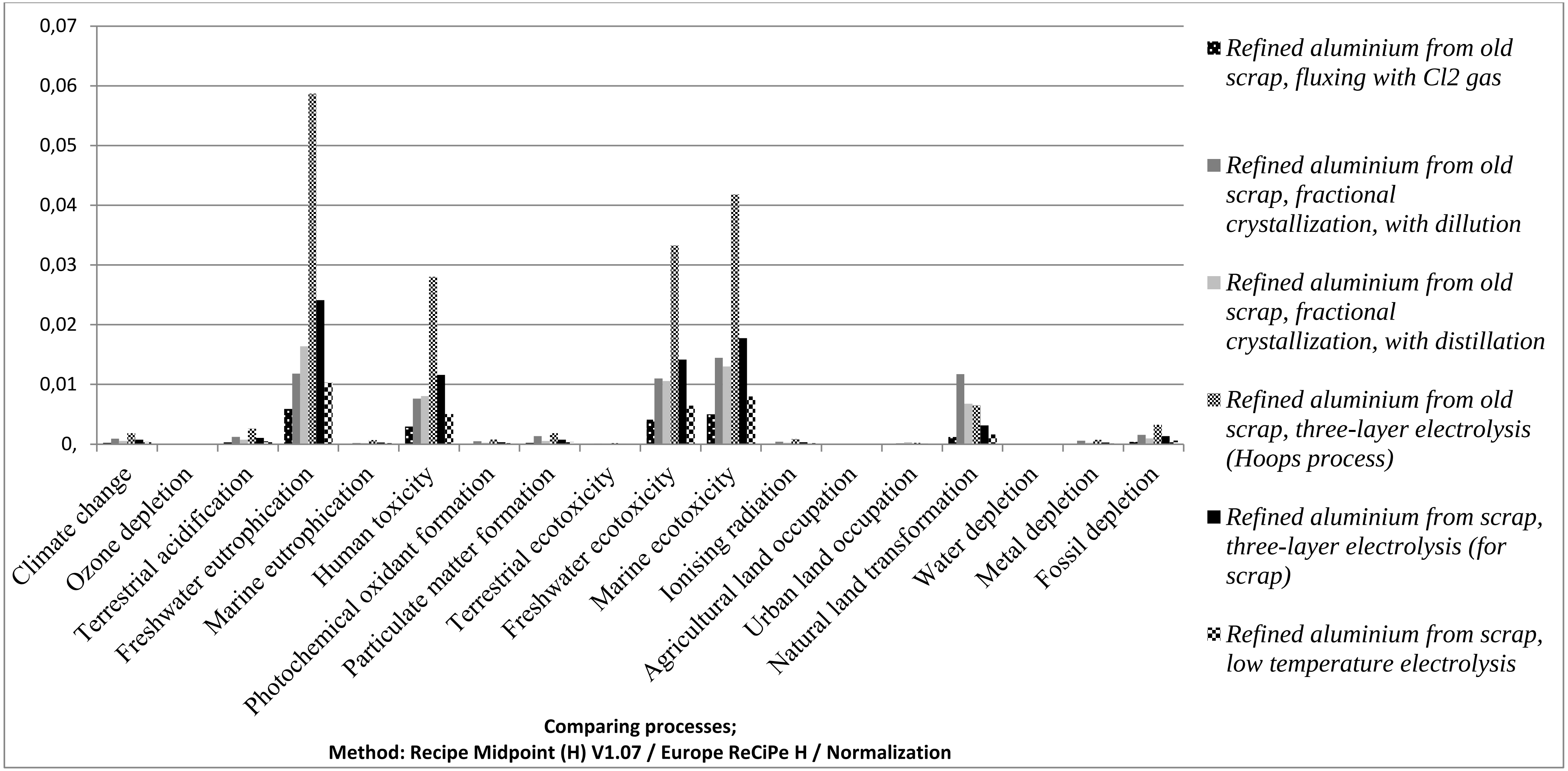


Figure 30: Normalised results for comparison between the six production scenarios, based on midpoint indicators. This figure indicates how much each indicator is affected by the various production scenarios.

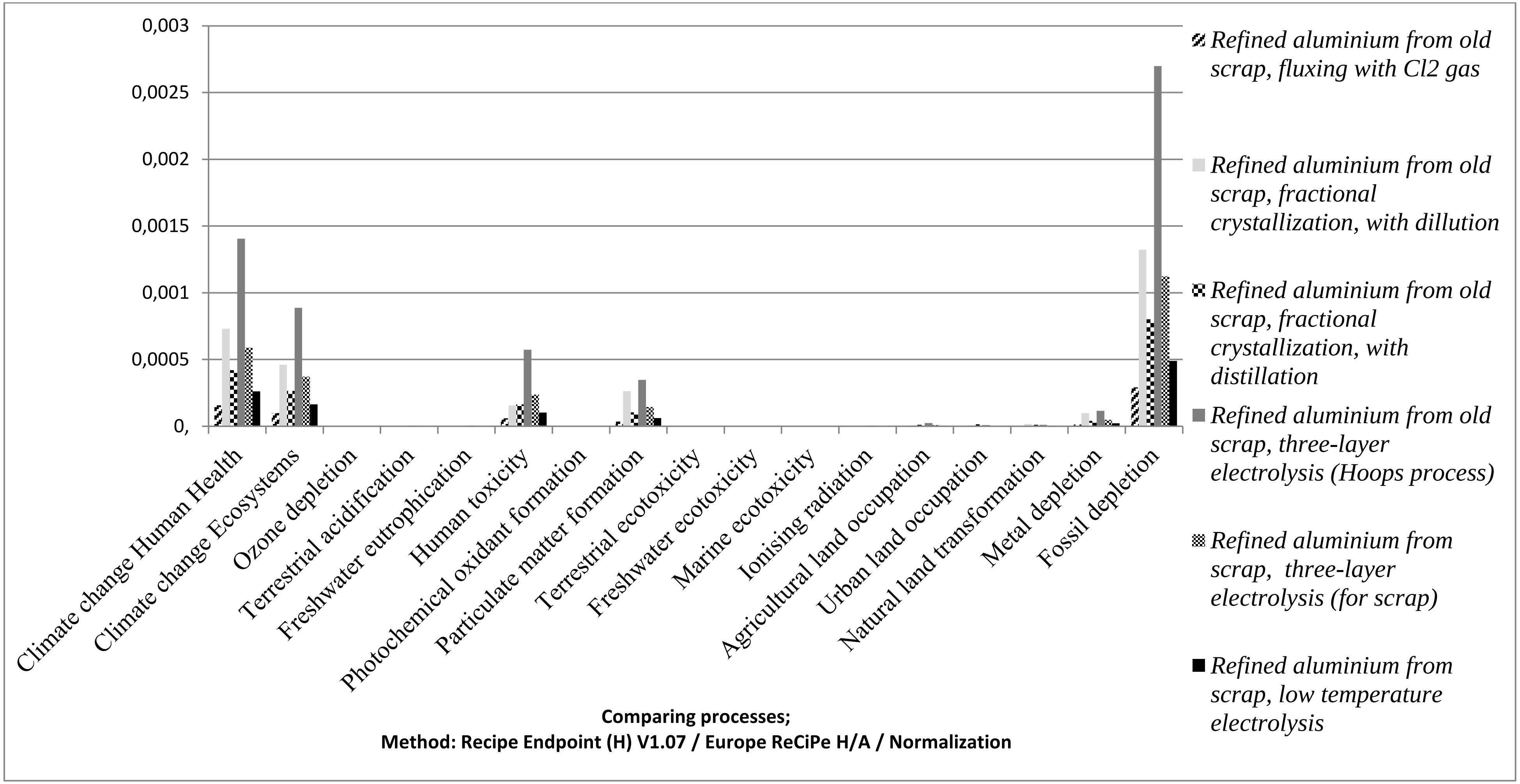


Figure 31: Normalised results for comparison between the six production scenarios, based on endpoint indicators. This figure indicates how much each indicator is affected by the various production scenarios.

6. Discussion

Results

The results from this assessment clearly show a relation between the energy requirement of a process, and the environmental burden associated with the same process. An important note here is that energy use is the main inventory for all the processes as well. This is related to two things. First, energy use is the only issue accounted for, for many of the processes. The other issue is that the most of the chemicals used are capable of being reused, and therefore the environmental burdens related to them are spread out.

There are several uncertainties related to when these result will be relevant. It is mentioned in the introduction that the use of aluminium is expected to continue its increase, and that this eventually will cause cascade recycling to be outdated. It is possible that this will be delayed compared to current projections, for example due to a discovery of more areas where low purity aluminium can be utilised. The long lifetime of aluminium products must also be carefully considered. The lifetime might also increase for future products and will also delay the need to refine post-consumer scrap back to high purity alloys.

Data availability

This is a general issue related to LCA and unfortunately establishing good inventory for the relevant refining processes proved very difficult. Energy requirements for all the processes were to some extent available through thermodynamic calculations, but this can only supply indications. Actual energy use is hard to determine, and results can only be as good as the numbers used in the calculations. It seems that since cascade recycling has not proved any problems so far, that little effort has been put on exploring possibilities for recycling of scrap aluminium. However the realisation that cascade recycling might become problematic in the future seems to have begun to enter the minds of industry and a selected number of studies have been conducted with an aim to refine scrap (Boender et al., 2002, Kahveci and Unal, 2000, Kevorkijan, 2010a, Le Brun et al., 2007, Schwarz and Wendt, 1995). As Sillekens et al. (2000) states; *"a shift towards closed loop recycling is unavoidable"*. But there are many different aluminium alloys presently used, so the imagined scrap compositions that have posed as bases for the conducted investigations vary. This causes an issue with determining the plausibility that a method is entirely transferable to any scrap and if so how efficient is can be assumed to be.

Limitations of problem formulation

It is clear that the problem formulation restricts the obtainable results. The first of the three main objectives of this thesis was to identify possible refining technologies to be applied to remove excess alloying elements in a given scrap composition. The given scrap composition and the alloys that is set as a goal to achieve through refining are the main limiting aspects of the problem formulation. The experience and impression through the work with this thesis is that the excess alloying elements that it is desirable to remove from this case scrap are the hardest ones to remove. It is also a question as to how relevant it is to want to remove large amounts of for example Si. It might be more realistic to assume that scrap containing high levels of Si would be recycled to alloys allowing higher levels of Si as Si is one of the most pervasive elements to remove. A very limiting aspect of the problem limitation is that the limits for Cu and Zn were set to be zero. This is very unrealistic and close to impossible to obtain. It is very worth mentioning that if these limits were adjusted that fractional crystallisation would be the preferred refining method. This method has the ability to be adjusted according to the desirable purity, and has very low energy use compared to all other refining methods applied. Fluxing is also irrelevant for the given scrap composition and the given limits that have to be met after refining. However this is a method which is well established and used by industry. The results for this process show low energy use, but has some negative impacts due to the use of chemicals causing hazardous waste. Sillekens (200) clearly state that the yield of fractional crystallisation as refining method is strongly dependent on the scrap composition it is used on, and which purity limits it is desirable to achieve. An important effect of scrap composition is the interaction between the present tramp elements which can make them harder or easier to remove.

Validity of results

Based on lack of good available data, and the limitations set by the problem formulation, it can be discussed how valid the results obtained in this thesis are. However, since the trend that energy use holds such a large responsibility of the total impacts caused by all scenarios, some conclusions can be drawn. Another conclusion which can be drawn from this is the motive to encourage improved sorting processes to separate for example cast alloys from wrought alloys to avoid extensive refining. So even though no definite statement can be made solely based on the results obtained in this life cycle assessment, there are some important issues that are addressed and can be used as a basis for further work.

Future solutions

As the introduction states, this thesis wanted to investigate future possibilities to refine scrap. The data used to explore this are based on present technologies both related to the refining processes and related to energy production. This can give a basis for understanding the possibilities that lies for future options, but it cannot be completely established to how these technologies will look in the future. It is interesting to reflect on how the results can be interpreted with a thought of future application. With a much cleaner energy production mix, the yield of each process might become more important. If clean electricity becomes cheaper it might get interesting to introduce refining of scrap earlier. If products are designed to recycled refining might become redundant all together.

Economic aspects

It is stated in the introduction that the main perspective of this thesis is the environmental perspective. But one must have in mind that it is mainly economics that control which technologies are implemented by industry. The added value to scrap must be higher than the cost of any refining process to make refining economically preferable. A recommendation based solely from an environmental perspective might be presumed inadequate. The wish is to be able to give recommendations on which refining processes should be investigated further and serve as possible industrialised solutions for future refining of post-consumer aluminium scrap. To do this some economic aspects must be considered. The impression from this work is that electrolysis is in general very expensive, because the use of inorganic salts causes extra costs due to disposal and cleaning of the chemicals used. This is also the most energy demanding processes, and energy is costly. These costs together must be weighed against the added value to the scrap to be able to give complete recommendations. An impressions obtained during the work with this thesis is that a better sorting process which make fractional crystallisation sufficient as refining method might be a good compromise between economic and environmental aspects.

Sorting of scrap versus refining of scrap

The last issue which must be discussed is the balance between sorting and refining of the scrap. It is evident that careful sorting will simplify the refining which has to take place considerably. So, when is it better to use resources on sorting carefully and the re-melt and reuse the alloys as they are? For careful sorting to be profitable considerable amounts of scrap should be locally available since the collection stage might be costly and make it difficult to provide stable sources of scrap. EAA (2004) mention that remelters are getting more specialised, this might indicate that remelters are no longer only handling locally available scrap but also importing scrap suitable for their reining methods. It is a plausible future scenario that such stable sources might be available, based on the assumptions that the use of aluminium will continue to increase in the future.

Closed loop recycling of aluminium

This is being done for some product groups of aluminium, i.e. beverage cans. Or, it is not entirely closed knowing that beverage cans consist of multiple alloys which cannot all be recycled back to their origins. To establish or build an industry on closed loop recycling of aluminium alloys, different options should be explored. Kevorkijan (2010b) explores with the thought of developing more recycling friendly aluminium alloys. This means alloys which can be used in a variety of products which are collected together for recycling at the end of their lifetime. An uncertainty related to recycling of scrap is the unsecure supply of raw-material. If use of aluminium continues to increase, it can be assumed that this flow of supply get more stable and it might get easier to establish foundries based on this. It is clear that from an environmental perspective a closed-loop recycling is preferable as this requires few or no refining process to take place. It is difficult to predict when this would be a feasible solution. A recommendation is to shift the train of thought from “how to recycle scrap back to existing alloys” to “how create alloys that are better suited for recycling”. This is of course only relevant for future productions, since there is a large amount of already produces alloys out there already in use. But it would be interesting to investigate whether or not and how to move the general focus over to how to produce more recyclable products/alloys.

7. Conclusion

The aim of this study was to identify possible solutions for future refining of secondary aluminium, and to do an environmental assessment of these refining methods. The results show that the main impacts associated with each of the identified refining possibilities are due to energy use. Therefore a conclusion must be that refining methods such as low temperature electrolysis and fractional solidification are preferable from an environmental perspective. The best alternative would be to use fractional crystallisation, without any additional refining steps, since this is the refining method requiring the smallest amount of energy. Possible improvements might be to move from non-renewable energy sources to towards the use of more renewable energy sources.

An overall impression based on this work is that it might a large potential in improving the sorting of post-consumer scrap. It was found for all scenarios that this process is only responsible for a small share of the total impacts to all impact categories. Compared to the energy used during the refining steps the energy used during sorting is very low. So an alternative improvement is to put more effort into this step to avoid energy requiring refining.

Another possibility is to develop alloys or products which are made with a motive to be recycled. This can be done by for example broadening the use of existing alloys or possibly merging alloys with similar qualities.

But some important “statements” can be confirmed from the results though. Kevorkijan, amongst others, state the importance of trying to close the loop of products or similar alloys. This will ease the refining process quite a lot and most likely be more environmentally sound. It has been clearly

To sum up the conclusion, the main contributions from this thesis must be that;

1) To be able to improve the refining methods and the scrap investigated in this analysis, the energy production must be improved. It was found that electricity use is the main issue for all scenarios, and since the energy mix used is largely based on fossil fuels, this will have a large impact on for example climate change.
2) The second issues it to underline the effect/importance of closing product or alloy loops. This might be easier for some product groups and alloy compositions
3) A third issue is to move the focus from which refining is possible, to how to avoid refining. There might be a large potential in the sorting step of the production route investigated for this thesis, as this uses very little energy compared to the other steps.
4) The last suggestion is the development of new alloys and product which are more fit to be recycled. Either by broadening the use of existing alloys, or maybe merging some alloys together to ease recycling. Doing this will ease the closing of product/alloy loops and help avoid energy requiring refining steps.

8. References

ASHTARI, P., DAVIS, B. & SADAYAPPAN, K. 2012a. Removal of Si from recycled aluminium alloys. *International Journal of Cast Metals Research,* 25**,** 100-102.

ASHTARI, P., TETLEY-GERARD, K. & SADAYAPPAN, K. 2012b. Removal of iron from recycled aluminium alloys. *Canadian Metallurgical Quarterly,* 51**,** 75-80.

BOENDER, W., WARINGA, C. J., KRIELAART, G. P., FOLKERTSMA, A. & VERDOES, D. Refining aluminium scrap with fractional crystallisation: Assessing its feasibility with thermodynamics. *In:* PETERSON, R., ed., 2002 Seattle, WA. 1061-1068.

BOIN, U. M. J. & BERTRAM, M. 2005. Melting standardized aluminum scrap: A mass balance model for Europe. *JOM,* 57**,** 26-33.

CHEN, C., WANG, J., SHU, D. & SUN, B. 2011. Removal of iron impurity from aluminum by electroslag refining. *Materials Transactions,* 52**,** 1320-1323.

CLASSEN, M., ALTHAUS, H. J., BLASER, S., TUCHSCHMID, M., JUNGBLUTH, N., DOKA, G., FAIST EMMENGGER, M. & SCHARNHORST, W. 2009. Life cycle inventories of Metals. Final report ecoinvent data v2.1, No 10. Empa Dübendorf: Swiss centre of Life Cycle Inventories, Dübendorf, CH, Online-Version under: www.ecoinvent.ch.

DAVIS, J. R. 1993. *Aluminum and aluminum alloys,* ASM international.

DAWLESS, R. K. & GRAZIANO, R. E. 1981. Fractional crystallization process. Aluminum Company of America, Pittsburg, Pa: United States Patent.

DAWLESS, R. K., TROUP, R. L., MEIER, D. L. & ROHATGI, A. 1988. Production of extreme-purity aluminum and silicon by fractional crystallization processing. *Journal of Crystal Growth,* 89**,** 68-74.

DEWAN, M. A., RHAMDHANI, M. A., MITCHELL, J. B., DAVIDSON, C. J., BROOKS, G. A., EASTON, M. & GRANDFIELD, J. F. 2011. Control and removal of impurities from Al melts: A review. Melbourne, VIC.

EAA 2004. Aluminium recycling: the road to high quality products. *European Aluminium Association (EEA), Brussels*.

ENGH, T. A. 1992. Principles of metal refining. *Oxford University Press, Walton St, Oxford OX 2 6 DP, UK, 1992. 473*.

EUROPEAN COMMISSION 2010. ILCD Handbook: General guide for LCA- detailed guidance. . *In:* JOINT RESEARCH CENTRE - INSTITUTE FOR ENVIRONMENT AND SUSTAINABILITY (ed.). (EUR 24708 EN) Luxemburg: Publications Office of the European Union, Available at; http://lct.jrc.ec.europa.eu/pdf-directory/ILCD-Handbook-General-guide-for-LCA-DETAIL-online-12March2010.pdf.

EUROPEAN COMMISSION. 2012. *Understanding the Seventh Framework Programme* [Online]. Available: http://ec.europa.eu/research/fp7/index_en.cfm?pg=understanding [Accessed 16.04 2013].

FJELD, A., EDUSSURIYA, S., EVANS, J. W. & MUKHOPADHYAY, A. Mathematical modeling of the chlorine fluxing of aluminum. *In:* KVANDE, H., ed., 2005 San Francisco, CA. 963-968.

FJELD, A., EVANS, J. W. & CHESONIS, D. C. Gas fluxing of aluminum: Comparison of computational fluid dynamics models and experiments. 2006 San Antonio, TX. 771-776.

GAO, J. W., SHU, D., WANG, J. & SUN, B. D. 2009. Study on iron purification from aluminium melt by Na 2B 4O 7 flux. *Materials Science and Technology,* 25**,** 619-624.

GAUSTAD, G., OLIVETTI, E. & KIRCHAIN, R. 2012. Improving aluminum recycling: A survey of sorting and impurity removal technologies. *Resources, Conservation and Recycling,* 58**,** 79-87.

GESING, A., AUBUCHON, B., TOREK, P., DALTON, R. & WOLANSKI, R. Assuring continued recyclability of automotive aluminum alloys: Chemical-composition-based sorting of wrought and cast Al shred. *In:* DAS, S. K., ed., 2003 San Diego, CA. 3-14.

GESING, A. J. 2005. Aluminium, recycling and transportation. Gesing Consultants.

GULBRANDSEN-DAHL, S., HENNING, R. M., JENSRUD, O., ØSTHUS, R., SØRBØ, S. & TOMOVIC-PETROVIC, S. 2013. Silicon content limitation level on formability and product performance in cast and formed AlMgSi. Norway: SINTEF Raufoss Manufacturing (for SuPLight EU FP7 project 263302).

HOBERG, H., WOLF, S. & MEIER-KORTWIG, J. Modelling the material flow of recycling processes for aluminium alloys by means of technical recycling quotas. *In:* GABALLAH, I., HAGER, J. & SOLOZABAL, R., eds., 1999 San Sebastian. 1023-1033.

ISO 14040:2006 2006. ISO 14040:2006 *Environmental management - Life cycle assessment - Principles and framework.* Geneva Switzerland: ISO copyright office.

ISO 14044:2006 2006. ISO 14044:2006. *Environmental management - Life cycle assessment - Requirements and guidelines.* Geneva Switzerland: ISO copyright office.

KAHVECI, A. I. & UNAL, A. Refining of a 5XXX series aluminum alloy scrap by Alcoa fractional crystallization process. *In:* STEWART JR, D. L., DALEY, J. C. & STEPHENS, R. L., eds., 2000. 979-991.

KAMAVARAM, V. & REDDY, R. G. Physical and thermal properties of ionic liquids used in aluminum electrorefining at low temperatures. *In:* DAS, S. K., ed., 2003 San Diego, CA. 299-307.

KEVORKIJAN, V. 2002. The recycle of wrought aluminum alloys in Europe. *JOM,* 54**,** 38-41.

KEVORKIJAN, V. 2010a. Advanced recycling of wrought aluminium alloys. *Metall,* 64**,** 414-418.

KEVORKIJAN, V. 2010b. The recycling of standard quality wrought aluminum alloys from low-grade contaminated scrap. *JOM,* 62**,** 37-42.

KIRCHAIN, R. & COSQUER, A. 2007. Strategies for maintaining light metal reuse: Insights from modeling of firm-wide raw materials availability and demand. *Resources, Conservation and Recycling,* 51**,** 367-396.

KJOS, O. S., SOLHEIM, A., SKYBAKMOEN, E., GUDBRANDSEN, H., ROLSETH, S., BERGSTRØM, T. & MARTINEZ, A. M. 2011. Elektrokjemisk raffinering av Al-skrap (Prosjektnr. 805484). *In:* SINTEF MATERIALER OG KJEMI (ed.).

LE BRUN, P., KRÄUTLEIN, C., ROMBACH, G., POULY, P., DE VRIES, P. & LUYTEN, J. Removal of intermetallic particles for the purification of aluminum alloys. 2007 Orlando, FL. 657-664.

MAJIDI, O., SHABESTARI, S. & ABOUTALEBI, M. 2007. Study of fluxing temperature in molten aluminum refining process. *Journal of materials processing technology,* 182**,** 450-455.

MODARESI, R. & MÜLLER, D. B. 2012. The role of automobiles for the future of aluminum recycling. *Environmental Science and Technology,* 46**,** 8587-8594.

NAKAJIMA, K., TAKEDA, O., MIKI, T., MATSUBAE, K., NAKAMURA, S. & NAGASAKA, T. 2010. Thermodynamic analysis of contamination by alloying elements in aluminum recycling. *Environmental Science and Technology,* 44**,** 5594-5600.

PETRUCCI, R. H. 2011. *General chemistry: principles and modern applications*, Toronto, Pearson Canada.

QIAN, F. U., DONG, X. U. & EVANS, J. W. 1998. Chlorine fluxing for removal of magnesium from molten aluminum part I. Laboratory-scale measurements of reaction rates and bubble behavior. *Metallurgical and Materials Transactions B: Process Metallurgy and Materials Processing Science,* 29**,** 971-978.

QIAN, F. U. & EVANS, J. W. 1998. Chlorine fluxing for removal of magnesium from molten aluminum Part II. Mathematical model. *Metallurgical and Materials Transactions B: Process Metallurgy and Materials Processing Science,* 29**,** 979-986.

ROMBACH, G. Future availability of aluminium scrap. LIGHT METALS-WARRENDALE-PROCEEDINGS-, 2002. TMS, 1011-1018.

SCHLESINGER, M. E. 2007. *Aluminum recycling*, Boca Raton, Fla., CRC Press.

SCHWARZ, V. & WENDT, H. 1995. Electrorefining of aluminium scrap from chloride melts. *Journal of Applied Electrochemistry,* 25**,** 34-40.

SILLEKENS, W. H., SCHADE VAN WESTRUM, J. A. F. M., BRUINSMA, O. S. L., MEHMETAJ, B. & NIENOORD, M. Refining aluminium scrap by means of fractional crystallisation: Basic experimental investigations. *In:* STEWART JR, D. L., DALEY, J. C. & STEPHENS, R. L., eds., 2000. 963-977.

SILLEKENS, W. H., VERDOES, D. & BOENDER, W. 2002. Refining aluminium scrap by means of fractional crystallisation: Status and prospects for development. *Metall,* 56**,** 468-473.

STRØMMAN, A. H. 2010. *Methodological Essential of LCA (Compendium for course TEP4223 at NTNU)*, NTNU, Trondheim.

STULL, D. R. & PROPHET, H. 1971. JANAF thermochemical tables. *In:* US DEPARTMENT OF COMMERCRE, N. B. O. S. (ed.). Washington, D.C.: DTIC Document.

SUPLIGHT. 2013. *suplight* [Online]. Available: http://www.suplight-eu.org/ [Accessed 16.04 2013].

TAN, R. B. H. & KHOO, H. H. 2005. An LCA study of a primary aluminum supply chain. *Journal of Cleaner Production,* 13**,** 607-618.

UTIGARD, T. 1998. The properties and uses of fluxes in molten aluminum processing. *JOM Journal of the Minerals, Metals and Materials Society,* 50**,** 38-43.

VERDOES, D. & VISSCHER, H. 2001. Method and device for separating metals and/or metal alloys of different melting points. Delft, NL: United States Patent.

WEI, W. 2012. Energy consumption and carbon footprint of secondary aluminium cast house (Master thesis). Royal Institute of Technology, Sweden: Department of Materials Science and Engineering, Division of Energy and Furnace Technology.

WORLD ALUMINIUM. 2013. *Primary aluminium production* [Online]. Available: http://www.world-aluminium.org/statistics/ [Accessed 01.06 2013].

ZAPP, P., ROMBACH, G. & KUCKSHINRICHS, W. The future of automotive aluminium. *In:* PETERSON, R., ed., 2002 Seattle, WA. 1003-1010.

ZHANG, L., LV, X., TORGERSON, A. T. & LONG, M. 2011. Removal of impurity elements from molten aluminum: A review. *Mineral Processing and Extractive Metallurgy Review,* 32**,** 150-228.

ZUMDAHL, S. S. 2005. *Chemical principles*, Boston, New York, Houghton Mifflin Company

Appendix A – Selected properties of relevant elements

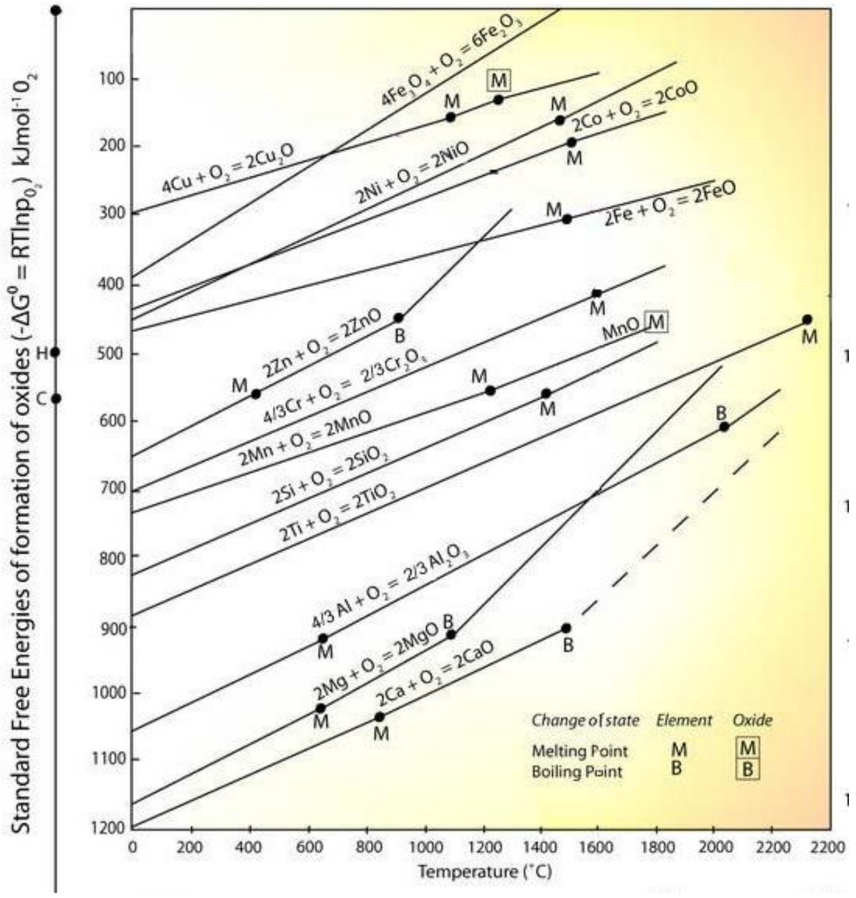
Selected properties of relevant elements

	Al	Si	Mg	Fe	Cu	Mn	Zn
Melting point	933 K	1685 K	922 K	1009 K	1356 K	1518 K	692 K
Boiling point	2766 K	3513 K	1378 K	3145 K	2848 K	2235 K	1180 K
Distribution coefficient*	-	0.1	0.25	0.03	0.14	0.93	0.5
Distribution coefficient**	-	0.13	0.45	0.03	0.17	0.62	0.87

* This distribution coefficient is based on Kahveci and Unal (2000)

** This distribution coefficient is based on Sillekens et al. (2000)

Ellingham diagram



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Appendix B - Energy calculations

This appendix includes all the energy calculations conducted in relation to this thesis. Thermodynamic values are all based on JANAF Thermodynamic Tables (Stull and Prophet, 1971). The reference state used for these tables is atmospheric pressure and a temperature of 298.15K.

Symbols and terminology

H: Enthalpy

Tm: melting point

M_x: molar mass of substance x

T: Temperature, it is specified whether Kelvin [K] or degrees Celsius [°C] are used

(l): liquid phase

(s): solid (crystal) phase

C_p°: Heat capacity at constant pressure

Q: Heat

ΔH°_f = standard enthalpy of formation. This is the enthalpy associated with the reaction and forming of a given compound from its elements, with each substance in its thermodynamic standard state at the given temperature.

$\Delta H^\circ_m = \Delta H^\circ_{Tm}(l) - \Delta H^\circ_{Tm}(s)$: Enthalpy change during melting.

$Q = C_p \cdot \Delta T$: Determines heat per kg needed to raise the temperature by ΔT degrees.

Unit conversion

The unit for enthalpy used in the JANAF Tables are kcal/mole. A more general unit more suitable for the context of this thesis is kJ/kg, or kWh/kg.

$M_{Al} = 26.9815$ g/mol ≈ 0.02698 kg/mol

1kcal = 4.184 kJ

1 kWh = 3600 kJ

1 kJ ≈ 0.00028 kWh

Consequently; 1 kcal/mol$_{(Al)}$ = 1550.78 kJ/kg$_{(Al)}$ = 0.4342 kWh/kg$_{(Al)}$.

Relevant thermodynamic values

$\Delta H^\circ_m (Al_{(s)}) = 3970 \pm 77.5$ kJ/kg

$C_p^\circ (Al_{(l)}) = 1.177$ kJ/kg/K (this is constant for all temperatures)

Enthalpies for aluminium in its reference state at the given temperature;

$H°_{(900\ K)}$ - $H°_{(298K)}$ = 4.064 kcal/mole $\longrightarrow$ $\Delta H°_{(900K)}$ = 6302.37 kJ/kg

$H°_{(1000\ K)}$ - $H°_{(298K)}$ = 7.397 kcal/mole $\longrightarrow$ $\Delta H°_{(1000K)}$ = 11 471.12 kJ/kg

$H°_{(1100\ K)}$ - $H°_{(298K)}$ = 8.156 kcal/mole $\longrightarrow$ $\Delta H°_{(1100K)}$ = 12 648.16 kJ/kg

$H°_{(1200\ K)}$ - $H°_{(298K)}$ = 8.915 kcal/mole $\longrightarrow$ $\Delta H°_{(1200K)}$ = 13 825.20 kJ/kg

Energy required melting pure aluminium

$\Delta H°_m$ ($Al_{(s)}$) = 3970 kJ/kg ± 77.5 kJ/kg = 1.11 ± 0.02 kJ/kg kWh/kg

$\Delta H°_m$ ($Al_{(s)}$) is the energy used to transfer the aluminium from solid state to molten state at the melting point. The total energy need have to include the heating of the solid from the initial temperature up to the melting point as well. This is done by extrapolating the values

Extrapolating (melting point 660°C = 933 K):

(11 471.12 – 6302.3) / (1000-900) = ($\Delta H°_{(933K)}$ – 6302.3) / (933-900)

$\Delta H°_{(933K)}$ = 8008.01 kJ/kg = 2.24 kWh/kg

Energy needed to reach 740°C in the molten aluminium

740°C = 1013 K

Use extrapolation (assumption of linear slope) to identify the enthalpy change to reach this temperature:

(12 648.16 – 11 471.12) / (1100-1000) = ($\Delta H°_{(1013K)}$ – 11 471.12) / (1013-1000)

$\Delta H°_{(1013K)}$ = 11 624.14 kJ/kg = 3.25 kWh/kg

Energy needed to reach 900°C in the molten aluminium

900°C = 1173 K

Use extrapolation (assumption of linear slope) to identify the enthalpy change to reach this temperature:

(13 825.20 – 12 648.16) / (1200-1100) = ($\Delta H°_{(1173K)}$ – 12 648.16) / (1173-1100)

$\Delta H°_{(1173K)}$ = 13 507.40 kJ/kg = 3.78 kWh/kg

NOTE: These calculations are based on observations related to pure aluminium, and can thus only be conceived indicative for scrap compositions.

Appendix C – Results from impact assessment in tabular form

Table A: Results for scenario 1a – Hoopes process as refining method.

Impact category	**Unit**	**Total**	***Shredding and sorting, Aluminium***	***Melt aluminium scrap, induction furnace***	***Secondary casting***	***Three layer electrolysis (Hoopes process), old aluminium scrap***
Climate change	kg CO2 eq	2,03E+01	1,29E-01	8,04E-01	2,68E-01	1,91E+01
Ozone depletion	kg CFC-11 eq	5,26E-07	2,55E-09	2,05E-08	1,85E-08	4,84E-07
Human toxicity, cancer effects	CTUh	2,09E-06	1,01E-08	8,51E-08	2,99E-08	1,97E-06
Human toxicity, non-cancer effects	CTUh	1,48E-06	6,74E-09	5,86E-08	3,45E-08	1,38E-06
Particulate matter	kg PM2.5 eq	7,69E-03	3,01E-05	3,08E-04	4,16E-05	7,31E-03
Ionizing radiation HH	kg U235 eq	5,23E+00	1,98E-02	2,09E-01	3,61E-02	4,97E+00
Ionizing radiation E (interim)	CTUe	1,55E-05	5,89E-08	6,20E-07	1,12E-07	1,48E-05
Photochemical ozone formation	kg NMVOC eq	3,97E-02	1,68E-04	1,59E-03	3,43E-04	3,76E-02
Acidification	molc H+ eq	1,17E-01	4,47E-04	4,66E-03	5,57E-04	1,11E-01
Terrestrial eutrophication	molc N eq	1,37E-01	5,82E-04	5,51E-03	1,22E-03	1,30E-01
Freshwater eutrophication	kg P eq	2,43E-02	8,91E-05	9,75E-04	7,57E-05	2,32E-02
Marine eutrophication	kg N eq	1,76E-02	7,10E-05	6,94E-04	3,67E-04	1,65E-02
Freshwater ecotoxicity	CTUe	3,12E+01	2,59E-01	1,24E+00	7,97E-01	2,89E+01
Land use	kg C deficit	5,59E+00	3,44E-02	1,01E-01	8,38E-02	5,37E+00
Water resource depletion	m3 water eq	2,00E-02	8,89E-05	7,10E-04	2,42E-03	1,68E-02
Mineral, fossil & ren resource depletion	kg Sb eq	2,92E-05	1,35E-07	1,23E-06	3,61E-07	2,75E-05
Non renewable, fossil	MJ	2,23E+02	8,77E-01	8,85E+00	2,40E+00	2,10E+02
Renewable, water	MJ	3,07E+00	1,29E-02	1,22E-01	4,69E-02	2,88E+00
Renewable, wind, solar, geothe	MJ	8,92E-02	5,07E-04	3,35E-03	6,42E-03	7,90E-02
Renewable, biomass	MJ	3,93E+00	1,46E-02	1,58E-01	1,91E-02	3,73E+00
Non-renewable, biomass	MJ	5,46E-05	2,75E-07	3,60E-06	3,74E-06	4,70E-05
Non-renewable, nuclear	MJ	5,43E+01	2,05E-01	2,17E+00	3,67E-01	5,16E+01

Table B: Results for Hoopes process.

Impact category	**Unit**	**Total**	***European mix, electricity for SuPLight***	***Aluminium electrolysis, plant/RER/I U***	***Disposal, dross from Al electrolysis, 0% water, to residual material landfill/CH U***
Climate change	kg CO2 eq	1,81E+01	1,81E+01	2,52E-03	1,05E-06
Ozone depletion	kg CFC-11 eq	4,60E-07	4,60E-07	1,58E-10	3,82E-13
Human toxicity, cancer effects	CTUh	1,87E-06	1,87E-06	1,11E-09	4,23E-14
Human toxicity, non-cancer effects	CTUh	1,31E-06	1,31E-06	1,63E-09	5,09E-14
Particulate matter	kg PM2.5 eq	6,95E-03	6,94E-03	2,09E-06	7,25E-10
Ionizing radiation HH	kg U235 eq	4,72E+00	4,72E+00	4,57E-04	1,14E-07
Ionizing radiation E (interim)	CTUe	1,40E-05	1,40E-05	1,40E-09	3,48E-13
Photochemical ozone formation	kg NMVOC eq	3,57E-02	3,57E-02	9,86E-06	1,05E-08
Acidification	molc H+ eq	1,05E-01	1,05E-01	2,26E-05	7,80E-09
Terrestrial eutrophication	molc N eq	1,24E-01	1,24E-01	5,88E-05	3,40E-08
Freshwater eutrophication	kg P eq	2,20E-02	2,20E-02	2,69E-06	8,77E-11
Marine eutrophication	kg N eq	1,56E-02	1,56E-02	3,27E-06	2,26E-06
Freshwater ecotoxicity	CTUe	2,75E+01	2,75E+01	1,64E-02	6,52E-07
Land use	kg C deficit	5,10E+00	5,23E+00	-1,25E-01	3,39E-05
Water resource depletion	m3 water eq	1,60E-02	1,60E-02	3,95E-06	5,56E-09
Mineral, fossil & ren resource depletion	kg Sb eq	2,61E-05	2,60E-05	8,60E-08	8,73E-12
Non renewable, fossil	MJ	2,00E+02	2,00E+02	2,96E-02	3,35E-05
Renewable, water	MJ	2,74E+00	2,74E+00	2,00E-03	2,10E-07
Renewable, wind, solar, geothe	MJ	7,50E-02	7,50E-02	5,19E-05	9,54E-09
Renewable, biomass	MJ	3,55E+00	3,54E+00	5,00E-03	4,68E-08
Non-renewable, biomass	MJ	4,47E-05	4,36E-05	1,06E-06	5,05E-11
Non-renewable, nuclear	MJ	4,90E+01	4,90E+01	4,79E-03	1,28E-06

Table C: Results for scenario 1b – adjusted three-layered electrolysis as refining method.

Impact category	**Unit**	**Total**	**Shredding and sorting, Aluminium**	**Melt aluminium scrap, induction furnace**	**Secondary casting**	**Three layer electrolysis, for scrap, old aluminium scrap**
Climate change	kg CO2 eq	8,49E+00	1,29E-01	8,04E-01	2,68E-01	7,29E+00
Ozone depletion	kg CFC-11 eq	2,27E-07	2,55E-09	2,05E-08	1,85E-08	1,85E-07
Human toxicity, cancer effects	CTUh	8,78E-07	1,01E-08	8,51E-08	2,99E-08	7,53E-07
Human toxicity, non-cancer effects	CTUh	6,27E-07	6,74E-09	5,86E-08	3,45E-08	5,27E-07
Particulate matter	kg PM2.5 eq	3,18E-03	3,01E-05	3,08E-04	4,16E-05	2,80E-03
Ionizing radiation HH	kg U235 eq	2,16E+00	1,98E-02	2,09E-01	3,61E-02	1,90E+00
Ionizing radiation E (interim)	CTUe	6,43E-06	5,89E-08	6,20E-07	1,12E-07	5,64E-06
Photochemical ozone formation	kg NMVOC eq	1,65E-02	1,68E-04	1,59E-03	3,43E-04	1,44E-02
Acidification	molc H+ eq	4,81E-02	4,47E-04	4,66E-03	5,57E-04	4,24E-02
Terrestrial eutrophication	molc N eq	5,71E-02	5,82E-04	5,51E-03	1,22E-03	4,98E-02
Freshwater eutrophication	kg P eq	1,00E-02	8,91E-05	9,75E-04	7,57E-05	8,87E-03
Marine eutrophication	kg N eq	7,43E-03	7,10E-05	6,94E-04	3,67E-04	6,30E-03
Freshwater ecotoxicity	CTUe	1,34E+01	2,59E-01	1,24E+00	7,97E-01	1,11E+01
Land use	kg C deficit	2,19E+00	3,44E-02	1,01E-01	8,38E-02	1,97E+00
Water resource depletion	m3 water eq	9,66E-03	8,89E-05	7,10E-04	2,42E-03	6,44E-03
Mineral, fossil & ren resource depletion	kg Sb eq	1,23E-05	1,35E-07	1,23E-06	3,61E-07	1,06E-05
Non renewable, fossil	MJ	9,26E+01	8,77E-01	8,85E+00	2,40E+00	8,05E+01
Renewable, water	MJ	1,29E+00	1,29E-02	1,22E-01	4,69E-02	1,10E+00
Renewable, wind, solar, geothe	MJ	4,05E-02	5,07E-04	3,35E-03	6,42E-03	3,02E-02
Renewable, biomass	MJ	1,62E+00	1,46E-02	1,58E-01	1,91E-02	1,43E+00
Non-renewable, biomass	MJ	2,63E-05	2,75E-07	3,60E-06	3,74E-06	1,87E-05
Non-renewable, nuclear	MJ	2,25E+01	2,05E-01	2,17E+00	3,67E-01	1,97E+01

Table D: Results for adjusted three-layered electrolysis.

Impact category	Unit	Total	European mix, electricity for SuPLight	Aluminium electrolysis, plant/RER/I U	Disposal, dross from Al electrolysis, 0% water, to residual material landfill/CH U
Climate change	kg CO2 eq	6,93E+00	6,92E+00	2,52E-03	1,05E-06
Ozone depletion	kg CFC-11 eq	1,76E-07	1,76E-07	1,58E-10	3,82E-13
Human toxicity, cancer effects	CTUh	7,15E-07	7,14E-07	1,11E-09	4,23E-14
Human toxicity, non-cancer effects	CTUh	5,01E-07	4,99E-07	1,63E-09	5,09E-14
Particulate matter	kg PM2.5 eq	2,66E-03	2,65E-03	2,09E-06	7,25E-10
Ionizing radiation HH	kg U235 eq	1,80E+00	1,80E+00	4,57E-04	1,14E-07
Ionizing radiation E (interim)	CTUe	5,36E-06	5,36E-06	1,40E-09	3,48E-13
Photochemical ozone formation	kg NMVOC eq	1,36E-02	1,36E-02	9,86E-06	1,05E-08
Acidification	molc H+ eq	4,03E-02	4,03E-02	2,26E-05	7,80E-09
Terrestrial eutrophication	molc N eq	4,73E-02	4,72E-02	5,88E-05	3,40E-08
Freshwater eutrophication	kg P eq	8,43E-03	8,42E-03	2,69E-06	8,77E-11
Marine eutrophication	kg N eq	5,98E-03	5,98E-03	3,27E-06	2,26E-06
Freshwater ecotoxicity	CTUe	1,05E+01	1,05E+01	1,64E-02	6,52E-07
Land use	kg C deficit	1,87E+00	2,00E+00	-1,25E-01	3,39E-05
Water resource depletion	m3 water eq	6,12E-03	6,11E-03	3,95E-06	5,56E-09
Mineral, fossil & ren resource depletion	kg Sb eq	1,00E-05	9,96E-06	8,60E-08	8,73E-12
Non renewable, fossil	MJ	7,65E+01	7,64E+01	2,96E-02	3,35E-05
Renewable, water	MJ	1,05E+00	1,05E+00	2,00E-03	2,10E-07
Renewable, wind, solar, geothe	MJ	2,87E-02	2,87E-02	5,19E-05	9,54E-09
Renewable, biomass	MJ	1,36E+00	1,35E+00	5,00E-03	4,68E-08
Non-renewable, biomass	MJ	1,77E-05	1,67E-05	1,06E-06	5,05E-11
Non-renewable, nuclear	MJ	1,87E+01	1,87E+01	4,79E-03	1,28E-06

Table E: Results for scenario 2 – low temperature electrolysis as refining method.

Impact category	**Unit**	**Total**	**Shredding and sorting, Aluminium**	**Secondary casting**	**Low temperature electrolysis, old aluminium scrap**
Climate change	kg CO2 eq	3,76E+00	1,27E-01	2,68E-01	3,37E+00
Ozone depletion	kg CFC-11 eq	1,07E-07	2,51E-09	1,85E-08	8,56E-08
Human toxicity, cancer effects	CTUh	3,88E-07	9,92E-09	2,99E-08	3,48E-07
Human toxicity, non-cancer effects	CTUh	2,86E-07	6,61E-09	3,45E-08	2,44E-07
Particulate matter	kg PM2.5 eq	1,36E-03	2,95E-05	4,16E-05	1,29E-03
Ionizing radiation HH	kg U235 eq	9,33E-01	1,94E-02	3,61E-02	8,77E-01
Ionizing radiation E (interim)	CTUe	2,77E-06	5,78E-08	1,12E-07	2,60E-06
Photochemical ozone formation	kg NMVOC eq	7,14E-03	1,65E-04	3,43E-04	6,64E-03
Acidification	molc H+ eq	2,06E-02	4,38E-04	5,57E-04	1,96E-02
Terrestrial eutrophication	molc N eq	2,48E-02	5,71E-04	1,22E-03	2,30E-02
Freshwater eutrophication	kg P eq	4,26E-03	8,74E-05	7,57E-05	4,09E-03
Marine eutrophication	kg N eq	3,35E-03	6,97E-05	3,67E-04	2,91E-03
Freshwater ecotoxicity	CTUe	6,17E+00	2,54E-01	7,97E-01	5,12E+00
Land use	kg C deficit	9,57E-01	3,37E-02	8,38E-02	8,39E-01
Water resource depletion	m3 water eq	5,48E-03	8,72E-05	2,42E-03	2,97E-03
Mineral, fossil & ren resource depletion	kg Sb eq	5,42E-06	1,33E-07	3,61E-07	4,93E-06
Non renewable, fossil	MJ	4,04E+01	8,61E-01	2,40E+00	3,72E+01
Renewable, water	MJ	5,70E-01	1,27E-02	4,69E-02	5,11E-01
Renewable, wind, solar, geothe	MJ	2,09E-02	4,98E-04	6,42E-03	1,40E-02
Renewable, biomass	MJ	6,97E-01	1,44E-02	1,91E-02	6,63E-01
Non-renewable, biomass	MJ	1,32E-05	2,70E-07	3,74E-06	9,22E-06
Non-renewable, nuclear	MJ	9,68E+00	2,02E-01	3,67E-01	9,11E+00

Table F: Results for low temperature electrolysis.

Impact category	**Unit**	**Total**	**European mix, electricity for SuPLight**	**Aluminium electrolysis, plant/RER/I U**	**Disposal, dross from Al electrolysis, 0% water, to residual material landfill/CH U**
Climate change	kg CO2 eq	3,20E+00	3,20E+00	2,52E-03	1,05E-06
Ozone depletion	kg CFC-11 eq	8,13E-08	8,12E-08	1,58E-10	3,82E-13
Human toxicity, cancer effects	CTUh	3,31E-07	3,30E-07	1,11E-09	4,23E-14
Human toxicity, non-cancer effects	CTUh	2,32E-07	2,31E-07	1,63E-09	5,09E-14
Particulate matter	kg PM2.5 eq	1,23E-03	1,23E-03	2,09E-06	7,25E-10
Ionizing radiation HH	kg U235 eq	8,33E-01	8,33E-01	4,57E-04	1,14E-07
Ionizing radiation E (interim)	CTUe	2,47E-06	2,47E-06	1,40E-09	3,48E-13
Photochemical ozone formation	kg NMVOC eq	6,30E-03	6,29E-03	9,86E-06	1,05E-08
Acidification	molc H+ eq	1,86E-02	1,86E-02	2,26E-05	7,80E-09
Terrestrial eutrophication	molc N eq	2,19E-02	2,18E-02	5,88E-05	3,40E-08
Freshwater eutrophication	kg P eq	3,89E-03	3,89E-03	2,69E-06	8,77E-11
Marine eutrophication	kg N eq	2,76E-03	2,76E-03	3,27E-06	2,26E-06
Freshwater ecotoxicity	CTUe	4,86E+00	4,85E+00	1,64E-02	6,52E-07
Land use	kg C deficit	7,97E-01	9,22E-01	-1,25E-01	3,39E-05
Water resource depletion	m3 water eq	2,82E-03	2,82E-03	3,95E-06	5,56E-09
Mineral, fossil & ren resource depletion	kg Sb eq	4,68E-06	4,60E-06	8,60E-08	8,73E-12
Non renewable, fossil	MJ	3,53E+01	3,53E+01	2,96E-02	3,35E-05
Renewable, water	MJ	4,85E-01	4,83E-01	2,00E-03	2,10E-07
Renewable, wind, solar, geothe	MJ	1,33E-02	1,32E-02	5,19E-05	9,54E-09
Renewable, biomass	MJ	6,30E-01	6,25E-01	5,00E-03	4,68E-08
Non-renewable, biomass	MJ	8,76E-06	7,70E-06	1,06E-06	5,05E-11
Non-renewable, nuclear	MJ	8,66E+00	8,65E+00	4,79E-03	1,28E-06

Table G: Results for scenario 3 – fractional crystallisation and dilution as refining method.

Impact category	**Unit**	**Total**	**Shredding and sorting, Aluminium**	**Melt aluminium scrap, induction furnace**	**Secondary casting**	**Fractional crystallization, melted old aluminium scrap**	**Aluminium, primary, at plant/RER U**
Climate change	kg CO2 eq	1,05E+01	4,92E-02	3,06E-01	2,68E-01	1,26E-01	9,79E+00
Ozone depletion	kg CFC-11 eq	6,28E-07	9,74E-10	7,83E-09	1,85E-08	5,45E-09	5,95E-07
Human toxicity, cancer effects	CTUh	2,75E-06	3,85E-09	3,24E-08	2,99E-08	1,47E-08	2,67E-06
Human toxicity, non-cancer effects	CTUh	6,51E-07	2,57E-09	2,23E-08	3,45E-08	1,84E-08	5,74E-07
Particulate matter	kg PM2.5 eq	5,48E-03	1,15E-05	1,17E-04	4,16E-05	5,72E-05	5,25E-03
Ionizing radiation HH	kg U235 eq	2,68E+00	7,55E-03	7,95E-02	3,61E-02	3,18E-02	2,53E+00
Ionizing radiation E (interim)	CTUe	8,01E-06	2,25E-08	2,36E-07	1,12E-07	9,47E-08	7,54E-06
Photochemical ozone formation	kg NMVOC eq	2,22E-02	6,39E-05	6,04E-04	3,43E-04	3,42E-04	2,08E-02
Acidification	molc H+ eq	5,59E-02	1,70E-04	1,78E-03	5,57E-04	8,32E-04	5,25E-02
Terrestrial eutrophication	molc N eq	7,58E-02	2,22E-04	2,10E-03	1,22E-03	1,53E-03	7,07E-02
Freshwater eutrophication	kg P eq	4,89E-03	3,40E-05	3,71E-04	7,57E-05	1,35E-04	4,28E-03
Marine eutrophication	kg N eq	7,77E-03	2,71E-05	2,64E-04	3,67E-04	1,28E-04	6,99E-03
Freshwater ecotoxicity	CTUe	3,11E+01	9,86E-02	4,73E-01	7,97E-01	2,16E-01	2,95E+01
Land use	kg C deficit	1,00E+01	1,31E-02	3,87E-02	8,38E-02	7,02E-03	9,86E+00
Water resource depletion	m3 water eq	8,89E-03	3,39E-05	2,71E-04	2,42E-03	1,37E-04	6,03E-03
Mineral, fossil & ren resource depletion	kg Sb eq	5,05E-05	5,16E-08	4,69E-07	3,61E-07	1,11E-06	4,85E-05
Non renewable, fossil	MJ	1,09E+02	3,34E-01	3,37E+00	2,40E+00	1,50E+00	1,02E+02
Renewable, water	MJ	2,67E+01	4,93E-03	4,66E-02	4,69E-02	2,75E-02	2,65E+01
Renewable, wind, solar, geothe	MJ	9,00E-02	1,93E-04	1,28E-03	6,42E-03	7,70E-04	8,14E-02
Renewable, biomass	MJ	7,53E-01	5,58E-03	6,02E-02	1,91E-02	5,93E-02	6,09E-01
Non-renewable, biomass	MJ	5,19E-04	1,05E-07	1,37E-06	3,74E-06	9,37E-07	5,12E-04
Non-renewable, nuclear	MJ	2,78E+01	7,83E-02	8,26E-01	3,67E-01	3,30E-01	2,62E+01

Table H: Results for fractional crystallisation.

Impact category	Unit	Total	European mix, electricity for SuPLight	Metal working factory/RER/I U
Climate change	kg CO2 eq	3,16E-01	2,66E-01	4,95E-02
Ozone depletion	kg CFC-11 eq	1,36E-08	6,77E-09	6,87E-09
Human toxicity, cancer effects	CTUh	3,68E-08	2,75E-08	9,29E-09
Human toxicity, non-cancer effects	CTUh	4,59E-08	1,92E-08	2,67E-08
Particulate matter	kg PM2.5 eq	1,43E-04	1,02E-04	4,09E-05
Ionizing radiation HH	kg U235 eq	7,94E-02	6,94E-02	1,00E-02
Ionizing radiation E (interim)	CTUe	2,37E-07	2,06E-07	3,05E-08
Photochemical ozone formation	kg NMVOC eq	8,55E-04	5,25E-04	3,30E-04
Acidification	molc H+ eq	2,08E-03	1,55E-03	5,32E-04
Terrestrial eutrophication	molc N eq	3,83E-03	1,82E-03	2,01E-03
Freshwater eutrophication	kg P eq	3,38E-04	3,24E-04	1,40E-05
Marine eutrophication	kg N eq	3,21E-04	2,30E-04	9,12E-05
Freshwater ecotoxicity	CTUe	5,39E-01	4,04E-01	1,35E-01
Land use	kg C deficit	1,76E-02	7,69E-02	-5,93E-02
Water resource depletion	m3 water eq	3,42E-04	2,35E-04	1,07E-04
Mineral, fossil & ren resource depletion	kg Sb eq	2,79E-06	3,83E-07	2,40E-06
Non renewable, fossil	MJ	3,74E+00	2,94E+00	8,02E-01
Renewable, water	MJ	6,87E-02	4,03E-02	2,85E-02
Renewable, wind, solar, geothe	MJ	1,92E-03	1,10E-03	8,22E-04
Renewable, biomass	MJ	1,48E-01	5,21E-02	9,61E-02
Non-renewable, biomass	MJ	2,34E-06	6,41E-07	1,70E-06
Non-renewable, nuclear	MJ	8,25E-01	7,21E-01	1,04E-01

Table I: Results for scenario 3b – fractional crystallisation and distillation as refining method.

Impact category	**Unit**	**Total**	**Shredding and sorting, Aluminium**	**Melt aluminium scrap, induction furnace**	**Fractional crystallization, melted old aluminium scrap**	**Secondary casting**	**Distillation, 900C, aluminium melt, old scrap**
Climate change	kg CO2 eq	6,06E+00	2,46E-01	1,53E+00	6,32E-01	2,68E-01	3,38E+00
Ozone depletion	kg CFC-11 eq	1,87E-07	4,87E-09	3,91E-08	2,73E-08	1,85E-08	9,71E-08
Human toxicity, cancer effects	CTUh	6,42E-07	1,93E-08	1,62E-07	7,35E-08	2,99E-08	3,57E-07
Human toxicity, non-cancer effects	CTUh	5,41E-07	1,28E-08	1,12E-07	9,18E-08	3,45E-08	2,90E-07
Particulate matter	kg PM2.5 eq	2,31E-03	5,73E-05	5,86E-04	2,86E-04	4,16E-05	1,34E-03
Ionizing radiation HH	kg U235 eq	1,51E+00	3,77E-02	3,98E-01	1,59E-01	3,61E-02	8,75E-01
Ionizing radiation E (interim)	CTUe	4,48E-06	1,12E-07	1,18E-06	4,73E-07	1,12E-07	2,60E-06
Photochemical ozone formation	kg NMVOC eq	1,25E-02	3,20E-04	3,02E-03	1,71E-03	3,43E-04	7,12E-03
Acidification	molc H+ eq	3,46E-02	8,52E-04	8,88E-03	4,16E-03	5,57E-04	2,01E-02
Terrestrial eutrophication	molc N eq	4,69E-02	1,11E-03	1,05E-02	7,66E-03	1,22E-03	2,64E-02
Freshwater eutrophication	kg P eq	6,80E-03	1,70E-04	1,86E-03	6,76E-04	7,57E-05	4,02E-03
Marine eutrophication	kg N eq	5,48E-03	1,35E-04	1,32E-03	6,42E-04	3,67E-04	3,02E-03
Freshwater ecotoxicity	CTUe	9,97E+00	4,93E-01	2,36E+00	1,08E+00	7,97E-01	5,24E+00
Land use	kg C deficit	1,21E+00	6,55E-02	1,93E-01	3,51E-02	8,38E-02	8,28E-01
Water resource depletion	m3 water eq	7,74E-03	1,69E-04	1,35E-03	6,85E-04	2,42E-03	3,11E-03
Mineral, fossil & ren resource depletion	kg Sb eq	1,81E-05	2,58E-07	2,35E-06	5,57E-06	3,61E-07	9,53E-06
Non renewable, fossil	MJ	6,62E+01	1,67E+00	1,69E+01	7,48E+00	2,40E+00	3,78E+01
Renewable, water	MJ	9,95E-01	2,46E-02	2,33E-01	1,37E-01	4,69E-02	5,53E-01
Renewable, wind, solar, geothe	MJ	3,28E-02	9,67E-04	6,38E-03	3,85E-03	6,42E-03	1,52E-02
Renewable, biomass	MJ	1,48E+00	2,79E-02	3,01E-01	2,96E-01	1,91E-02	8,34E-01
Non-renewable, biomass	MJ	2,71E-05	5,25E-07	6,85E-06	4,69E-06	3,74E-06	1,13E-05
Non-renewable, nuclear	MJ	1,56E+01	3,92E-01	4,13E+00	1,65E+00	3,67E-01	9,09E+00

Table J: Results for distillation.

Impact category	**Unit**	**Total**	**European mix, electricity for SuPLight**	**Metal working factory/RER/I U**
Climate change	kg CO2 eq	1,69E+00	1,64E+00	4,95E-02
Ozone depletion	kg CFC-11 eq	4,85E-08	4,17E-08	6,87E-09
Human toxicity, cancer effects	CTUh	1,78E-07	1,69E-07	9,29E-09
Human toxicity, non-cancer effects	CTUh	1,45E-07	1,18E-07	2,67E-08
Particulate matter	kg PM2.5 eq	6,70E-04	6,29E-04	4,09E-05
Ionizing radiation HH	kg U235 eq	4,37E-01	4,27E-01	1,00E-02
Ionizing radiation E (interim)	CTUe	1,30E-06	1,27E-06	3,05E-08
Photochemical ozone formation	kg NMVOC eq	3,56E-03	3,23E-03	3,30E-04
Acidification	molc H+ eq	1,01E-02	9,54E-03	5,32E-04
Terrestrial eutrophication	molc N eq	1,32E-02	1,12E-02	2,01E-03
Freshwater eutrophication	kg P eq	2,01E-03	2,00E-03	1,40E-05
Marine eutrophication	kg N eq	1,51E-03	1,42E-03	9,12E-05
Freshwater ecotoxicity	CTUe	2,62E+00	2,49E+00	1,35E-01
Land use	kg C deficit	4,14E-01	4,73E-01	-5,93E-02
Water resource depletion	m3 water eq	1,56E-03	1,45E-03	1,07E-04
Mineral, fossil & ren resource depletion	kg Sb eq	4,76E-06	2,36E-06	2,40E-06
Non renewable, fossil	MJ	1,89E+01	1,81E+01	8,02E-01
Renewable, water	MJ	2,76E-01	2,48E-01	2,85E-02
Renewable, wind, solar, geothe	MJ	7,61E-03	6,79E-03	8,22E-04
Renewable, biomass	MJ	4,17E-01	3,21E-01	9,61E-02
Non-renewable, biomass	MJ	5,65E-06	3,95E-06	1,70E-06
Non-renewable, nuclear	MJ	4,54E+00	4,44E+00	1,04E-01

Table K: Results for scenario 4 – fluxing with Cl_2 gas as refining method.

Impact category	**Unit**	**Total**	**Shredding and sorting, Aluminium**	**Melt aluminium scrap, induction furnace**	**Fluxing with Cl2 gas, aluminium melt**	**Secondary casting**
Climate change	kg CO2 eq	2,28E+00	1,24E-01	7,69E-01	1,11E+00	2,68E-01
Ozone depletion	kg CFC-11 eq	1,20E-07	2,46E-09	1,97E-08	7,89E-08	1,85E-08
Human toxicity, cancer effects	CTUh	2,36E-07	9,73E-09	8,15E-08	1,14E-07	2,99E-08
Human toxicity, non-cancer effects	CTUh	1,92E-07	6,48E-09	5,61E-08	9,52E-08	3,45E-08
Particulate matter	kg PM2.5 eq	7,94E-04	2,89E-05	2,95E-04	4,29E-04	4,16E-05
Ionizing radiation HH	kg U235 eq	5,60E-01	1,91E-02	2,00E-01	3,05E-01	3,61E-02
Ionizing radiation E (interim)	CTUe	1,67E-06	5,67E-08	5,93E-07	9,08E-07	1,12E-07
Photochemical ozone formation	kg NMVOC eq	4,22E-03	1,61E-04	1,52E-03	2,20E-03	3,43E-04
Acidification	molc H+ eq	1,19E-02	4,30E-04	4,46E-03	6,47E-03	5,57E-04
Terrestrial eutrophication	molc N eq	1,47E-02	5,60E-04	5,27E-03	7,64E-03	1,22E-03
Freshwater eutrophication	kg P eq	2,44E-03	8,57E-05	9,33E-04	1,35E-03	7,57E-05
Marine eutrophication	kg N eq	2,06E-03	6,84E-05	6,64E-04	9,64E-04	3,67E-04
Freshwater ecotoxicity	CTUe	3,92E+00	2,49E-01	1,19E+00	1,69E+00	7,97E-01
Land use	kg C deficit	5,44E-01	3,31E-02	9,71E-02	3,30E-01	8,38E-02
Water resource depletion	m3 water eq	4,21E-03	8,56E-05	6,80E-04	1,02E-03	2,42E-03
Mineral, fossil & ren resource depletion	kg Sb eq	3,42E-06	1,30E-07	1,18E-06	1,75E-06	3,61E-07
Non renewable, fossil	MJ	2,40E+01	8,44E-01	8,47E+00	1,23E+01	2,40E+00
Renewable, water	MJ	3,68E-01	1,24E-02	1,17E-01	1,91E-01	4,69E-02
Renewable, wind, solar, geothe	MJ	1,90E-02	4,88E-04	3,21E-03	8,84E-03	6,42E-03
Renewable, biomass	MJ	4,06E-01	1,41E-02	1,51E-01	2,21E-01	1,91E-02
Non-renewable, biomass	MJ	1,03E-05	2,65E-07	3,44E-06	2,85E-06	3,74E-06
Non-renewable, nuclear	MJ	5,80E+00	1,98E-01	2,08E+00	3,16E+00	3,67E-01

Table L: Results for fluxing with Cl_2 gas.

Impact category	**Unit**	**Total**	**Chlorine, liquid, production mix, at plant/RER U**	**European mix, electricity for SuPLight**	**Disposal, hazardous waste, 0% water, to underground deposit/DE U**
Climate change	kg CO2 eq	1,11E+00	3,17E-02	1,08E+00	5,40E-06
Ozone depletion	kg CFC-11 eq	7,86E-08	5,12E-08	2,73E-08	1,48E-13
Human toxicity, cancer effects	CTUh	1,14E-07	2,97E-09	1,11E-07	1,30E-12
Human toxicity, non-cancer effects	CTUh	9,47E-08	1,71E-08	7,76E-08	5,44E-13
Particulate matter	kg PM2.5 eq	4,27E-04	1,42E-05	4,12E-04	2,92E-09
Ionizing radiation HH	kg U235 eq	3,03E-01	2,29E-02	2,80E-01	5,03E-07
Ionizing radiation E (interim)	CTUe	9,04E-07	7,09E-08	8,33E-07	1,55E-12
Photochemical ozone formation	kg NMVOC eq	2,19E-03	7,15E-05	2,12E-03	1,55E-08
Acidification	molc H+ eq	6,44E-03	1,83E-04	6,26E-03	2,43E-08
Terrestrial eutrophication	molc N eq	7,60E-03	2,59E-04	7,34E-03	5,42E-08
Freshwater eutrophication	kg P eq	1,34E-03	3,11E-05	1,31E-03	3,04E-09
Marine eutrophication	kg N eq	9,60E-04	3,06E-05	9,29E-04	5,27E-09
Freshwater ecotoxicity	CTUe	1,68E+00	4,78E-02	1,63E+00	1,45E-05
Land use	kg C deficit	3,28E-01	1,77E-02	3,11E-01	6,53E-06
Water resource depletion	m3 water eq	1,02E-03	6,93E-05	9,50E-04	7,59E-09
Mineral, fossil & ren resource depletion	kg Sb eq	1,74E-06	1,91E-07	1,55E-06	1,40E-11
Non renewable, fossil	MJ	1,23E+01	3,82E-01	1,19E+01	7,90E-05
Renewable, water	MJ	1,90E-01	2,76E-02	1,63E-01	1,19E-06
Renewable, wind, solar, geothe	MJ	8,79E-03	4,34E-03	4,45E-03	9,14E-08
Renewable, biomass	MJ	2,20E-01	9,72E-03	2,10E-01	1,46E-05
Non-renewable, biomass	MJ	2,83E-06	2,43E-07	2,59E-06	6,64E-11
Non-renewable, nuclear	MJ	3,14E+00	2,32E-01	2,91E+00	6,12E-06