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Life cycle assessment of a novel production route for scandium recovery from bauxite residues

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A B S T R A C T

Scandium (Sc) has various technological applications, but the concentrations of Sc in ores are low. Both, the mining of low concentrated Sc and the production of industrial-grade Sc are a heavy burden on the environment. Bauxite residue (BR) from alumina production represents one of the major sources of Sc in Europe (Ochsenkühn-Petropulu et al., 1994). The goal of this study is to assess the environmental impacts from cradle to gate of a novel production route developed in the Scandium Aluminium Europe project (SCALE) to extract Sc at concentrations <100 ppm from BR, to concentrate and upgrade it to pure ScF_3 and Sc_2O_3 and ultimately to refine it to an aluminium scandium master alloy with 2 % Sc mass fraction (AlSc2 %). Results show that the global warming potential (GWP), measured in $CO_{2\text{-eq}}$ per kg Sc_2O_3, generated with the novel route is about half the GWP of the state-of-the-art Sc_2O_3 production from rare earth tailings when applying equal allocation principles. The initial process step to dissolve BR and extract Sc consumes elevated amounts of acid and energy and is responsible for at least 80 % of the route's total environmental impact. The amount of the generated filter cake (FC) is equal to the amount of the BR input and is a potential resource for cement clinker production. The ecotoxicological study indicates that both FC and BR are slightly ecotoxic.

1. Introduction

Scandium (Sc) is one of the most valuable elements in the periodic table and is usually grouped with rare earth elements (REEs) (Dukov, 2007). Sc has various technological applications. For example, the efficiency of solid fuel cells is improved by using scandium oxide (Sc_2O_3) to produce scandia-stabilised-zirconia (Ciacchi et al., 1991). Scandium trifluoride (ScF_3) is the only precursor used to produce scandium metal (Martinez et al., 2018). ScF_3 also enables AlSc alloy production by an electrometallurgical process such as the Hall–Héroult process (Scandium Aluminium Europe, 2021). High strength AlSc alloys can reduce aircraft weight and fuel consumption (Goran Djukanovic, 2017).

The wide range of technological applications coupled with the limited supply of Sc lead to its classification as a critical raw material for the European economy (Commission et al., 2020). The global supply and consumption of Sc is ~10 to 25 tonnes per year. Major Sc-producing countries are China (66 % of global Sc production), Russia (26 %) and Ukraine (7 %) (U.S. Geological Survey, 2021).

Sc is present in traces of ores exploited in aluminium, cobalt, iron, molybdenum, nickel, phosphate, tantalum, tin, titanium, tungsten, uranium, zinc and zirconium production. Commercially, Sc is mainly extracted as a co-product from REE, uranium mining or titanium dioxide production (Wang et al., 2011). The concentrations of Sc in ores are low. Both, the mining of low concentrated Sc and the production of industrial-grade Sc are a heavy burden on the environment. The currently applied production routes consume large amounts of water, energy, mineral acids and solvents, generating pollutant emissions in the water and air (EPA, 2012). To produce ScF_3 by state-of-the-art processes,

Abbreviations: ASC, Antisolvent crystallisation; BR, Bauxite residue; CAL, Calcination; FC, Filter cake(s); FP, Filter press; LMA, Leaching with mineral acid; PLS, Pregnant leach solution; RMR, Refining by metallothermic reduction; SCALE, Scandium Aluminium Europe project (EU Horizon 2020); SIR, Selective ion recovery and ion exchange; SX, Solvent extraction.

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Sc_2O_3 is fluorinated with gaseous hydrogen fluoride (HF), which is environmentally harmful and potentially dangerous for human health (Harata et al., 2008).

Acid tailings from REE-bearing ore processing are often not managed properly, thus leading to the contamination of water bodies and serious human health issues (Huang et al., 2016). The presence of naturally occurring radioactive materials, such as thorium in ores (EPA, 2012), can result in high concentrations of radioactive materials in products, by-products and wastes.

Bauxite residue (BR) from alumina production represents one of the major sources of Sc in Europe (Ochsenkühn-Petropulu et al., 1994). For example, Greek BR contains 70–100 ppm of Sc (Gamaletsos et al., 2019) (Scandium Aluminium Europe, 2021). 800,000 tonnes of BR generated in an aluminium plant in Greece contain 60–80 tonnes of Sc (Scandium Aluminium Europe, 2021).

Innovative technologies and new connections in the processing chain were recently developed in the Scandium Aluminium Europe project (SCALE), which extract Sc from BR with mineral acid leaching followed by selective ion recovery and ion exchange (Davris et al., 2021) to purify the obtained Sc-enriched solution with solvent extraction. With direct crystallisation and calcination, the obtained Sc strip solution is then processed to ScF_3 and Sc_2O_3, which are commercial products (Peters et al., 2019). Through the metallothermic reduction of Al with ScF_3, an aluminium scandium master alloy with 2 % Sc mass fraction (AlSc2 %) is produced. The novel process route bypasses existing expensive and, in the case of fluorination with gaseous HF. This study reports on (1) the environmental impacts of the potentially upscaled production of Sc products using BR as Sc source and on (2) the ecotoxicity of the filter cake from mineral acid leaching of BR. The study is based on experimental data from pilot testing conducted in the SCALE project.

2. Literature review

In this chapter, data from the literature review concerning established Sc extraction from (1) TiO_2 acid waste and (2) rare earth ore (REO) tailings are presented. Zhang et al. (2018) report LCA results for Sc_2O_3 obtained from acid waste of TiO_2 production in China using primary data. The study presents Global Warming Potential (GWP) values of 743 kg $CO_{2\text{-eq}}$ per kg of Sc_2O_3 and 1135 kg $CO_{2\text{-eq}}$ per kg of Sc metal and a total energy consumption of 3081.4 MJ per kg of Sc_2O_3 and 4715 MJ per kg of Sc metal.

Koltun & Tharumarajah (2014) use the price allocation model to estimate the environmental impact of a single REO extracted from an REO concentrate. The study applies an average concentration of 0.1 % of Sc_2O_3 in bastnäsite and monazite ores and a price of US$7200 per kg of Sc_2O_3 to allocate the environmental impact of all produced REO to Sc_2O_3. The U.S. Geological Survey (2021) reports a lower price of US $3900 per kg of Sc_2O_3. The LCI only considers energy and water consumption but none of the chemicals required to process the ore and its intermediate products. GWP values for Sc_2O_3 add up to ~5500 kg $CO_{2\text{-eq}}$ per kg. A much higher GWP can be assumed when the amount of chemicals reported by Talens Peiró & Villalba Méndez (2013) is considered in the LCI.

Wang et al. (2020) present an LCA study of Sc_2O_3 production by exploiting RE tailings in the Bayan Obo Mine in China. The authors of this study indicate that the economic allocation of environmental impacts to the different products used in most studies is not appropriate due to the high variability of Scandium prices and have therefore chosen mass allocation approach to allocate the environmental impacts to Sc_2O_3 and by-products (Fe concentrate, Fluorite coarse, RE-, Fe, S-, and Nb concentrate) at the different beneficiation and melting stages. As the mass share of Sc_2O_3 is rather low compared to the mass share of the by-products, a lower proportion of the environmental impact is consequently attributed to Sc_2O_3, while a higher proportion is attributed to the by-products. Consequently, the GWP of Sc_2O_3 value is 3940 kg $CO_{2\text{-eq}}$ per kg Sc_2O_3, which is around 30 % lower than the value presented by Koltun & Tharumarajah (2014), which have only considered water and energy consumption by using economic allocation. However, the GWP values presented in these two studies are higher than those of Sc_2O_3 of from acid waste of TiO_2 production presented by Zhang et al. (2018), for which fewer resources are needed, since the Sc source is already in an acidic solution.

3. Materials and methods

3.1. Process chain description

The novel production route uses BR as a Sc source. Sc will be extracted by 1) leaching with mineral acid (LMA) as described by Ochsenkuehn-Petropoulou et al. (2018) and Balomenos et al. (2021), followed by 2) selective ion recovery and ion exchange (SIR) according to Balomenos et al. (2021). The pilot plant used for LMA and SIR is described by Davris et al. (2021). It will then be purified with 3) solvent extraction (SX) similar to Hedwig et al. (2023). This is followed by 4) fractional antisolvent crystallisation (ASC) according to Peters et al. (2019), and 5) calcination (CAL) to produce ScF_3 and Sc_2O_3 as described by Kaya et al. (2018). Finally, the process chain is completed with 6) refining by metallothermic reduction (RMR) to produce a AlSc2 % master alloy similar to Brinkmann et al. (2019). See S1 in the supplementary information for a more detailed process chain description of novel Sc recovery using BR.

3.2. Life cycle assessment

Life cycle assessment (LCA) is a well-known, internationally recognised methodology to assess the potential environmental impacts of products, processes or services. Environmental hotspots can be identified and decision making for greening the business supported. The methodology essentially involves the gathering of material and energy flows of the up- and downstream processes of a system over the entire life cycle as well as the evaluation of environmental consequences. The LCA in this study is performed following the ISO14040:2006 Standard (ISO, 2006), which includes the following steps: (1) definition of goal and scope, (2) life cycle inventory (LCI), (3) life cycle impact assessment (LCIA) and (4) interpretation.

3.2.1. Goal and scope definition

The goal of this study is to assess the environmental burden from cradle to gate of a novel production route developed in the SCALE project to extract Sc at a concentration of < 100 ppm from BR, to concentrate and to upgrade it to a pure ScF_3 and Sc_2O_3 and to ultimately refine it to an AlSc2 % master alloy. Furthermore, the environmental burdens of Sc_2O_3 production of the novel route are compared with state-of-the-art production in the Bayan Obo Mine in China using the REE tailings as the Sc source (Wang et al., 2020).

The functional units (FU) in this study refer to the production of 1 kg of ScF_3, 1 kg of Sc_2O_3 and 1 kg of AlSc2 % master alloy, all obtained by the novel production process.

The system boundary includes the upstream processes of energy, chemicals and materials (considering transport) used in the production of Sc products. In the assessed scenario all process steps to produce Sc products are located at an alumina plant processing bauxite ore in the Bayer process. Furthermore, direct process emissions, such as CO_2 from neutralisation with limestone, diffuse volatile organic compounds (VOCs) and acid emissions, and upstream processes, such as the disposal of spent chemicals and process residues, are considered. The disposal or further use of the filter cake (FC) from LMA is beyond the system boundary. The FC amount from LMA is nearly equal to the BR amount from alumina production, which otherwise is mainly landfilled if Sc is not extracted. Furthermore, plant production facilities are not considered within the system boundary due to uncertainties of their LCIA impact.

3.2.2. Allocation rules

The output of LMA is the targeted PLS and the FC, a potential secondary resource in cement clinker production, which could replace bauxite as a source for Al_2O_3. Joyce & Björklund (2019) report on the potential use of BR form alumina production in cement clinker production. The FC is rich in aluminium, iron, calcium and silicon, but the amount of a pontential uptake at a cement clinker plant depends on the demand of these elements to balance the composition of the raw meal. The common approach to allocate the environmental impacts to the main- and by products follows the economic principle (Bailey et al., 2020). The uptake of the FC also depends on the transportation and landfilling costs of the FC at the alumina / scandium plant. Short transportation distances and elevated landfilling cost are seen as relevant factors to make the uptake of the FC economically attractive. For the calculation of the allocation factors the costs for landfilling the FC and producing the AlSc alloy were chosen. It is assumed that the landfilling cost of the FC is similar to the cost of landfilling BR which is about 9 US$ per ton (Ujaczki et al., 2018). The average price from 2016 to 2020 for AlSc alloy is 338 US$ per kg (U.S. Geological Survey, 2021). The allocation factor results from forming the ratio of the multiplication of the quantity with the price for an AlSc alloy and FC. Details are shown in Table S2 of the supplementary information. The allocation ratio of about 1:32 means that 97 % of the environmental impact should be allocated to the AlSc alloy, however, it was decided to allocate all impacts to the AlSc alloy because of large uncertainties regarding the uptake of FC in cement clinker in practice.

To make the LCIA results of this study comparable with Wang et al. (2020), a mass allocation approach was also applied for LMA, considering the mass ratio of PLS and FC (see Section 4.2).

3.2.3. Life cycle inventory

The LCI was modelled with SimaPro 9.0 LCA software. The amount of material and energy flows for the production processes are obtained from pilot test tests on LMA, SIR, SX, ASC CAL and RMR. The supply of materials and energy used in the processes, the generated emissions and the disposal of waste are modelled with background data and processes from the ecoinvent database version 3.5.

To predict material and energy flows close to industrial conditions, material and energy flow values were adjusted considering expert judgments. The material and energy balances to produce ScF_3, Sc_2O_3 and AlSc2 % are presented in Tables S3–S12 of the supplementary information. At LMA, which is the most energy and resource intensive process step, the measured energy consumption from the pilot tests is adjusted to 60 %. At SIR, it is assumed that the acidic filter cake is neutralised with $CaCO_3$. The generated CO_2 emissions are calculated stoichiometrically. To approximate closed loops for the use of NH_4F scrubbing solution and antisolvents for ASC, a recycling yield of 90 % is assumed. Utility inputs for solvent regeneration and factors to calculate emissions from BR, acids and solvents to air are estimated according to Geisler et al. (2004). The electricity consumption for the filter press is taken from the BREF document Common Waste Water and Waste Gas Treatment/Management Systems in the Chemical Sector (Brinkmann et al., 2016).

Fig. 1 shows a simplified flowchart of the SCALE AlSc2 % production route, with potential material flow loops, created in consultation with experts based on the results of pilot testing. The detailed material and energy balance to produce 1 kg of AlSc2 % is presented in Tables S3–S12 of the supplementary information. To produce 1 kg of AlSc2 %, ~1800 kg of BR (25 % wet) is required to obtain the Sc amount for the master alloy. The overall process yield adds up to ~25 %. The total energy input sums up to ~280 kWh / kg AlSc2 %. The inputs of chemicals (mainly H_2SO_4), primary aluminium and limestone sum up to ~490 kg / kg AlSc2 %. Assuming that all water for LMA and SIR is used in a closed loop, the freshwater consumption to compensate losses sums up to ~0.2 kg / kg AlSc2 %. The total amount of waste totals to ~1950 kg / kg AlSc2 %, but most of it can be used in the cement industry as secondary raw material. The total amount of final effluents sums up to ~340 kg / kg AlSc2 %, and the CO_2 emissions from neutralisation with limestone are ~40 kg / kg AlSc2 %.

The production route shown still has optimisation potential to reduce resource consumption. For example, a process-integrated solution for the reuse of the SIR raffinate is more desirable than the presented neutralisation with limestone. This would reduce not only the amount of SIR filter cake but also the production costs. Considering a potential industrial production at an annual processing of 800,000 tonnes of BR on a dry basis (1 million tonnes of BR on a 25 % wet basis), ~560 tonnes of

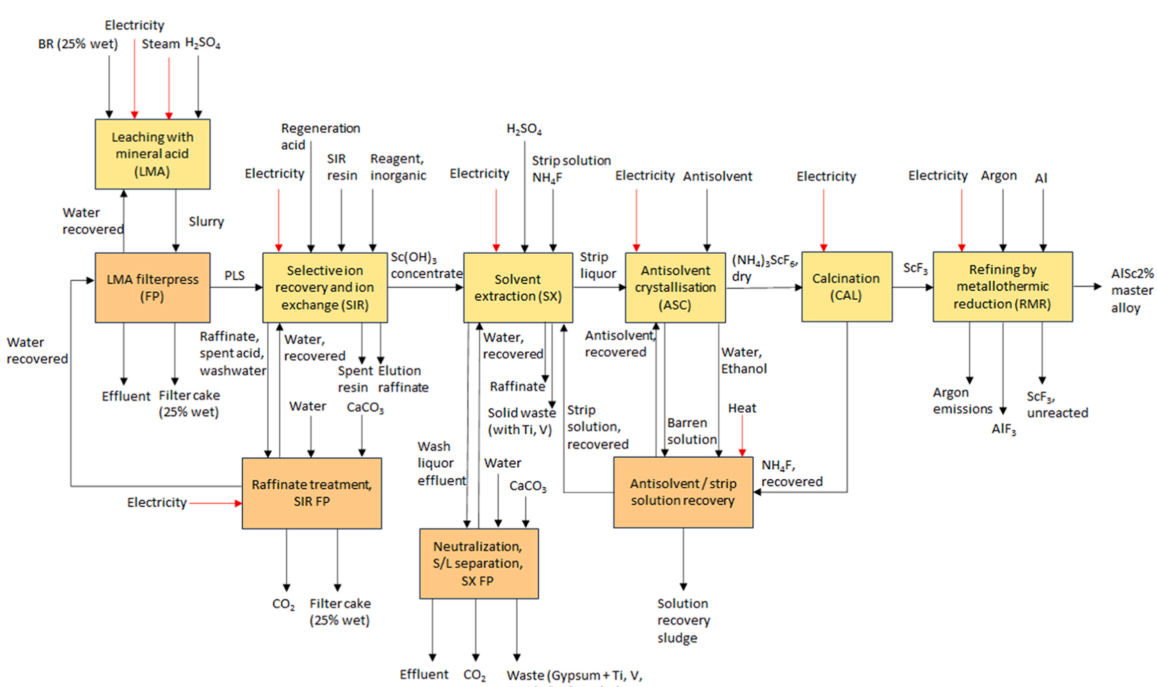

Fig. 1. Simplified flow chart of a potential SCALE production route created in consultation with experts based on the results of pilot testing.

AlSc2 % could be produced, and all BR outputs of the aluminium plant could further be processed, including the FC from LMA, which could be used for cement clinker production.

3.3. Life cycle impact assessment

In the LCIA phase, the environmental consequences of the inventory results will be evaluated in accordance with the functional units defined in Section 3.2.1. The inventory data are associated with environmental impact categories, such as climate change and natural resource depletion, which are supported by characterisation factors. When conducting an LCIA, the International Reference Life Cycle Data System (ILCD) Handbook (European Commission - Joint Research Centre, 2012) has recommended using the ILCD indicator, which includes 16 midpoint impact categories. As proposed by Sala et al. (2018), the weighting factors for each impact category were used to ensure that the focus is on the most relevant environmental aspects. The LCIA in this study refers to the ILCD indicators global warming potential (GWP) (Myhre et al., 2013) and cumulative energy demand (CED) (Frischknecht et al., 2007). Showing GWP and CED results separately enables the comparability with many others LCA studies using only these indicators. See Table S13 of the supplementary information for a detailed overview on the LCIA indicators.

3.4. Interpretation

In the last phase of an LCA, its results are interpreted in light of the goal and scope definition (e.g., by analysing impact contributions data uncertainty, parameter sensitivity and different evaluation models and as well as by comparing with other studies) to provide conclusions and recommendations (See chapter 5).

3.5. Ecotoxicity testing

Ecotoxicity testing of BR and LMA-FCs (see details in Table S14 in the supplementary information) was carried out with direct contact tests that applied a complex ecotoxicity test battery (Gruiz et al., 2015) (Pandard & Römbke, 2013), including testorganisms from three trophic levels for both water extracts (extracts made according to Pandard et al. [2006]) and solid samples. Genotoxicity assessment was also part of the test battery, as suggested by Römbke (2018), for the hazard assessment of wastes. The following tests were performed: *Aliivirbrio fischeri* (bacteria) bioluminescence inhibition (Leitgib et al., 2007)(Ujaczki et al., 2015); *Sinapis alba* (mustard plant) root and shoot elongation inhibition (Leitgib et al., 2007); *Daphnia magna* (crustacean, animal) mortality (Nagy et al., 2014), *Folsomia candida* (Collembola, animal) mortality tests (Leitgib et al., 2007)(Ujaczki et al., 2015); and SOS chromotest for genotoxicity testing (Environmental Bio-Detection Products Inc., 2016). Effective concentrations (EC20 and EC50, concentration causing 20/50 % inhibition) were calculated from the inhibition percentage (compared to an uncontaminated control) of a sample dilution series. EC20 values were considered the threshold dilution with tolerable toxic effect, and the samples were grouped into acute toxicity categories (I: no toxicity, II: slight acute toxicity) based on the EC50 values (Persoone et al., 2003). Samples were considered genotoxic if the induction factor was above 2 and the survival rate above 70 % (Environmental Bio-Detection Products Inc., 2016).

4. Results and discussion

4.1. Life cycle impact assessment

In this section, the LCIA of the SCALE products ScF_3, Sc_2O_3 and AlSc2 % master alloy is presented. Table S15 in the supplementary information provides a detailed overview on the environmental impacts of the products measured in the different ILCD impact categories. ScF_3 is the precursor to produce Sc_2O_3 and AlSc2 %; hence, the previous process steps LMA, SIR, SX, ASC and CAL are equal for all SCALE products. Sc_2O_3 is obtained by an additional calcination step of ScF_3 and the AlSc2 % master alloy by a metallothermic reduction of Sc_2O_3 with primary aluminium.[1] Furthermore, the results of the sensitivity analysis for Sc_2O_3 are discussed. In Table 1, CED (GJ/kg), total ILCD (mPt/kg) and GWP (kg CO_{2eq}/kg) values for the three SCALE products ScF_3, Sc_2O_3 and AlSc2 % are summarised.

The share of the impact categories (see section 3.2.4) on the total ILCD impact of the three products is almost equal. Freshwater ecotoxicity contributes 23–24 %; human toxicity, non-cancer effects, 19–20 %; human toxicity, cancer effects, 6–7 %; Mineral, fossil and renewable resource depletion, 18–19 %; climate change, 9 %; acidification, 7–8 %; water resource depletion, 2 %; and the remaining categories, 14–15 %. All ILCD impact values are presented in Table S15 of the supplementary information.

4.2. ScF_3 production

The environmental impact to produce 1 kg of ScF_3 (0.44 kg of Sc per kg of ScF_3) measured with the CED indicator sums up to almost ~100 GJ per kg of ScF_3. The ILCD indicator amounts to ~1190 mPt per kg of ScF_3. LMA contributes 93 % to the total ILCD impact, and the GWP value totals to ~3500 kg $CO_{2\text{-eq}}$ per kg of ScF_3.

4.3. Sc_2O_3 production

The environmental impact to produce 1 kg of Sc_2O_3 (0.65 kg of Sc per kg of Sc_2O_3) measured with the CED indicator sums up to ~150 GJ per kg of Sc_2O_3. The ILCD indicator amounts to ~1750 mPt per kg of Sc_2O_3. The GWP value is ~5300 kg $CO_{2\text{-eq}}$ per kg of ScF_3, and the CAL of ScF_3 to Sc_2O_3 adds less than 1 % to the total ILCD impact of the product.

4.4. AlSc2 % master alloy production

The Sc content in 1 kg AlSc2 % is lower by a factor of 22 and 32 than in 1 kg of ScF_3 and Sc_2O_3, respectively, which explains the correspondingly lower impact values of AlSc2 % production. The environmental impact to produce 1 kg of AlSc2 % master alloy measured with the CED indicator sums up to ~4.8 GJ per kg of AlSc2 %. Fig. 2 shows the environmental impact shares of process step inputs and emissions measured with the CED indicator in GJ per kg of AlSc2 % master alloy produced. LMA as the first process step has the dominant share of the total CED (~89 %) required in the AlSc2 % master alloy production chain. The CED of LMA consists of the environmental impact shares from the energy input for electricity[2] (european electricity mix) and steam[3] (total of ~1.4 GJ / kg AlSc2 %), and the use of sulfuric acid[4] (~2.8 GJ / kg AlSc2 %) based on ecoinvent database version 3.5. The CED from the energy input in the subsequent process steps totals ~0.3 GJ per kg of

Table 1
CED, total ILCD and GWP values for the three SCALE products ScF_3, Sc_2O_3 and AlSc2 %.

Impact measure	Unit	ScF_3	Sc_2O_3	AlSc2 %
CED	GJ/kg	99.8	148	4.8
Total ILCD	ILCD mPt/kg	1190	1750	59.2
GWP	kg $CO_{2\text{-eq}}$/kg	3500	5290	172

[1] Aluminium, primary, ingot {IAI Area, EU27 & EFTA} | production | Cut-off, U

[2] Electricity, medium voltage {RER} | market group for | Cut-off, U

[3] Steam, in chemical industry {RER} | production | Cut-off, U

[4] Sulfuric acid {RER} | market for sulfuric acid | Cut-off, U

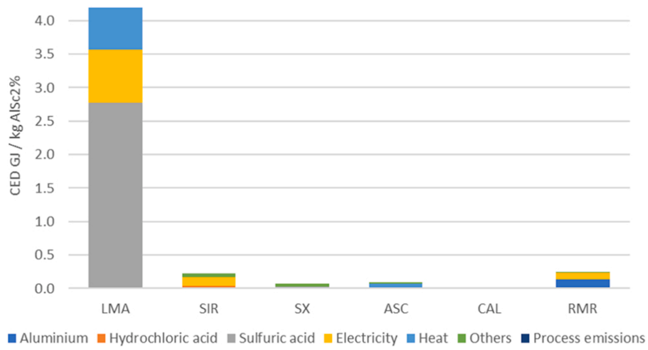

Fig. 2. Environmental impact shares of aluminium, chemicals, energy and other inputs, and process emissions measured with the CED indicator in GJ / kg AlSc2 % per process step. The CED value sums up to ~4.8 GJ per kg of SCALE AlSc2 % master alloy production.

AlSc2 %. Other smaller CED contributions from chemical use stem from the hydrochloric acid[5] input at SIR (~0.04 GJ / kg AlSc2 %) and the sulfuric acid input at SX (~0.03 GJ / kg AlSc2 %). The primary aluminium input at RMR corresponds to a CED of ~0.1 GJ per kg of AlSc2 % master alloy. Process emissions are not considered in the CED indicator.

The ILCD indicator amounts to ~59 mPt per kg of AlSc2 %. The categories 'water, mineral, fossil and renewable resource depletion', 'human toxicity' and 'climate change' contribute ~19 %, ~18 % and ~9 % to the total ILCD value, respectively. The main reasons for the high environmental impact in the 'climate change' category are the upstream energy production and the CO_2 emissions during the dissolution of limestone ($CaCO_3$)[6] particularly at SIR – and to a smaller extent, SX. The environmental impacts of the categories 'water, mineral, fossil and renewable resource depletion' and 'human toxicity' are mainly upstream of the SCALE process chain. Fig. 3 shows the environmental impact shares of process step inputs and emissions measured with the ILCD indicator in ILCD mPt per kg of AlSc2 % master alloy produced. As for the CED value, LMA also has the dominant share of the total ILCD value (~86 %) of the AlSc2 % master alloy production chain. The elevated amounts of sulfuric acid and energy consumption at LMA make ~74 % and ~9 % of the total ILCD impact of the production route, respectively. The primary aluminium input at RMR to produce an AlSc2 % master alloy contributes ~7 % to the total ILCD value. Emissions of the first four process steps LMA, SIR, SX and ASC in total make ~5 % of the total ILCD impact.

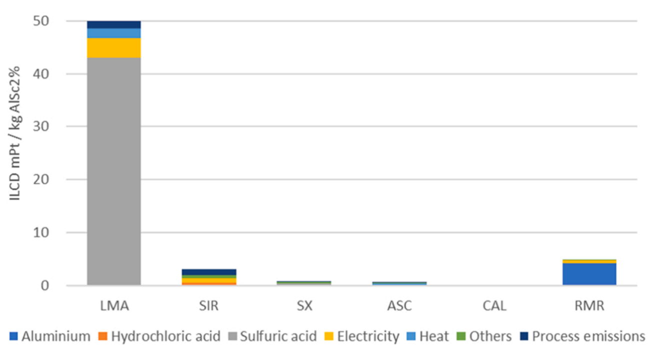

Fig. 3. Environmental impact shares of aluminium, chemicals, energy and other inputs as well as process emissions measured with the ILCD indicator in mPt / kg AlSc2 % per process step. The ILCD value sums up to ~59 mPt per kg of SCALE AlSc2 % master alloy production.

The GWP totals to ~170 kg $CO_{2\text{-eq}}$ per kg of AlSc2 %. The categories 'Ionising radiation' and 'freshwater ecotoxicity' contribute 5 % and 23 % to the total value, respectively. Fig. 4 shows the environmental impact shares of process step inputs and emissions measured with the GWP in kg $CO_{2\text{-eq}}$ per kg of AlSc2 % master alloy produced. As for the CED and ILCD values, LMA also has the dominant share of the GWP value (~63 %) of the AlSc2 % master alloy production chain. Sulfuric acid and energy consumption at LMA make ~25 % and ~38 % of the total ILCD impact of the production route, respectively. SIR as second process step also has a considerable share of the route's total GWP value (~27 %). SIR process emissions contribute ~21 % to the total ILCD value of the production chain. The primary aluminium input at RMR to produce an AlSc2 % master alloy comprises ~4 % of the total ILCD value.

The three indicators of environmental impact (i.e., CED, ILCD and GWP) all show similar shares of the individual process steps. LMA always has the highest share of environmental impact, while CAL has the lowest share. SIR, SX, ASC and RMR are between LMA and CAL for all indicators, with proportions always below ~5 %, with one exception: SIR has an environmental impact proportion of ~27 % considering the GWP, mainly due to the CO_2 emissions that occur during the neutralisation of the raffinate with limestone. For the CED and ILCD, the high impact of LMA is primarily due to the use of sulphuric acid, in contrast to the GWP, in which electricity outweighs the impact of sulphuric acid. Primary aluminium has the main share of environmental impact through RMR refining for all three indicators, with a contribution of ~3–7 % to the total environmental impact of AlSc2 % production.

4.5. *GWP comparison with traditional Sc_2O_3 extraction from REO tailings*

In this section, the GWP of the Sc_2O_3 production in the SCALE production route is compared with the GWP of traditional Sc_2O_3 extraction from REO tailings presented in Wang et al. (2020). Both production routes comprise resource intensive upstream processes compared to Sc extraction from acidic TiO_2 waste, for which fewer resources are needed, since the Sc source is already in an acidic solution.

The confidence limits of the characteristic value of GWP to produce Sc_2O_3 in the SCALE production route are determined through the LCA SimaPro software feature Monte Carlo simulation. Standard parameters have been used: confidence interval 95 %, and a random seed point to start the analysis.

Based on the economic allocation, all environmental impacts of the production are allocated to Sc products (see Section 3.2.2). The mean

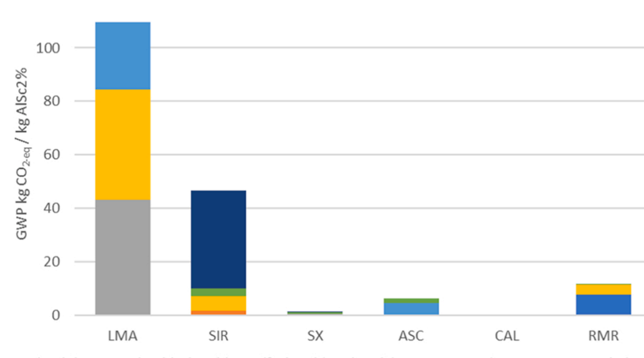

Fig. 4. Environmental impact shares of aluminium, chemicals, energy and other inputs as well as process emissions measured with the GWP in kg $CO_{2\text{-eq}}$ / kg AlSc2 % per process step. The GWP sums up to ~170 kg $CO_{2\text{-eq}}$ per kg of SCALE AlSc2 % master alloy production.

[5] Hydrochloric acid, without water, in 30 % solution state {RER} | market for | Cut-off, U

[6] Limestone, crushed, for mill {RoW} | market for limestone, crushed, for mill | Cut-off, U

value results in 5290 kg $CO_{2\text{-eq}}$/kg Sc_2O_3. At the 2.5 % and 97.5 % confidence interval, the GWP of Sc_2O_3 has a range of ~30 %. Details are shown in Table 2.

To compare the GWP values of Sc_2O_3 production from BR with the GWP values presented in Wang et al. (2020), an analysis of the mass-based allocation of GWP to the main Sc_2O_3 product and the FC by-product is carried out. The mass ratio of FC dry matter base (750 kg) to PLS (37 kg) is 20:1. Consequently, 5 % of the LMA environmental impact is allocated to the Sc-enriched PLS and 93 % to FC. The mass allocated GWP of BR-based Sc_2O_3 in the SCALE production route (1930 kg $CO_{2\text{-eq}}$ / kg Sc_2O_3) is about half the mass allocated GWP of Sc_2O_3 extracted from REE tailings (3940 kg $CO_{2\text{-eq}}$ / kg Sc_2O_3) indicated in the study of Wang et al. (2020).

4.6. LMA FC ecotoxicity results

Table 3 shows the ecotoxicity test results for LMA FC (from the pilot experiment), the primary waste stream from the process. For comparison, ecotoxicity data were included on the original BR. Water extracts model the potential ecotoxic effects of water leachates from LMA FC on the aquatic environment. Direct contact solid tests model the potential effects on the terrestrial ecosystem.

Most EC_{20} values are below EC_{20} values of five times sample dilutions (except for *D. magna* and *S. alba* direct contact tests), showing that this dilution would result in acceptable toxicity. *D. magna*, the aquatic crustacean, is the most sensitive test organism in this case for the water extracts of the samples. LMA FC is not toxic to the *F. candida* test organism. The original BR has similar ecotoxic properties. Ruyters et al, (2011) and Ujaczki et al, (2015) showed for other BRs that 5 % (20 times dilution) BR in soils had no significant toxic effect on selected test organisms (i.e., bacteria, plants and animals). Dauvin (2010) measured no toxic effect of the lixiviates or the solid fraction BR on *A. fischeri*. In contrast, Di Carlo et al, (2020) and Fourrier et al. (2021) found that different BRs had significant effects on the germination and development of *S. alba*, and the origin of the tested BR strongly modulated the toxic effect.

Based on the EC_{50} values, the LMA FC sample can be characterised as slightly toxic for both aquatic and terrestrial environments (Persoone et al., 2003). Therefore, when reused or disposed of properly, LMA FC would not pose a problem in subsurface waters. Slight toxicity may be due to the water-extractable Al content (226 mg/L) and the acidic pH (4.1) of the LMA FC. The water extractable Al content exceeds 100 times the European threshold values for groundwater (European Commission, 2009). Reducing the amount of sulphuric acid from 4 M to 1 M for Sc leaching from BR during the development of the technology resulted in the LMA FC falling into the same acute aquatic ecotoxicity category as the original BR (slight acute aquatic toxicity based on the median EC_{50} values). A detailed overview is shown in Table S16 in the supplementary information. The reduction of sulphuric acid use lessens the environmental impacts and costs.

The SOS chromotest shows that the FC water extract is not genotoxic at two times dilution (1 time dilution is cytotoxic for the *E. coli* bacterium; therefore, the results cannot be evaluated). The BR extract is not genotoxic either. Gelencsér et al, (2011) also found that the fugitive dust of BR was not genotoxic based on SOS Chromotest results. Dauvin, (2010) showed that marine discharged BR has no mutagenic effect based on the Ames reverse mutation test.

Based on the suggestions of Hennebert (2018), LMA FC is not considered ecotoxic for the aquatic environment according to the HP14 hazard category for wastes (EC_{50} thresholds for ecotoxic category: 6.3 times dilution for *A. fischeri* bacteria, 12.6 times dilution for *D. magna* crustacean, 14.2 times dilution for algae as primary producer). Based on the plant test, LMA FC is not hazardous for the terrestrial environment (threshold: 7.3 times dilution). Thus, the waste generated in the mineral acid leaching technology step can be categorised as non-toxic / slightly toxic and in the same ecotoxicity category as the original BR.

Table 2
GWP comparison of novel BR-based and traditional REE tailings-based production of Sc_2O_3.

Sc_2O_3 basis	Allocation	GWP (kg $CO_{2\text{-eq}}$ / kg Sc_2O_3)		
		Mean value	*Lower limit (2.5 %)*	*Upper limit (97.5 %)*
BR	Economic	5290	4590	6290
	Mass	1930	1860	2020
REE tailings (Wang et al., 2020)	Mass	3940	–	–

Table 3
Selected ecotoxicity test results for BR (original) and LMA FC (from the pilot experiment).

Sample	Test method	Effective concentration	BR	LMA FC PILOT
			x times dilution	
Water extract	*Aliivibrio fischeri* (bacteria) bioluminescence inhibition	EC_{20} (30 min)	<5.0	6.0
		EC_{50} (30 min)	<5.0	<5.0
	Sinapis alba (plant) shoot elongation	EC_{20} (72 h, shoot)	no inhibition at 1.0	6.0
		EC_{50} (72 h, shoot)		2.0
	Daphnia magna (animal) mortality	EC_{20} (48 h)	29.6	12.0
		EC_{50} (48 h)	9.0	10.8
	SOS Chromotest (genotoxicity)		not genotoxic at 1.0	not genotoxic at 2.0*
Direct contact with solid	*Sinapis alba* (plant) shoot inhibition	EC_{20} (shoot)	12.9	3.6
		EC_{50} (shoot)	2.7	2.0
	Folsomia candida (Collembola) (animal) mortality	EC_{20} (7 days)	2.6	no inhibition at 1.0
		EC_{50} (7 days)	1.5	

* Cytotoxic effect at 1.0 dilution

5. Conclusions

The novel production route developed in the SCALE project enables the valorisation of Sc content in BR. However, using LMA to dissolve elevated amounts of BR to extract low Sc content (<100 ppm) is resource and energy intensive and responsible for 90 % to the total ILCD impact of ScF_3 and Sc_2O_3 production and 80 % of the total ILCD impact of the AlSc2 % master alloy production. CO_2 emissions from the neutralisation step of the acidic waste at SIR with limestone have a considerable direct environmental impact on the production route. It is expected that after full integration of the Sc production route in an aluminium plant and process optimisation, limestone consumption can be reduced. Almost all other environmental impacts are associated with the provision of energy and chemicals, such as sulphuric acid for LMA. The amount of FC generated at LMA is about equal to the amount of BR input, and its concentrations of Al, Ca, Fe and Si allow a substation of primary raw materials in cement clinker production. Proximity of cement plants to aluminium plants (economic factor), elevated Fe concentration in the FC (technical factor) and high concentrations of radioactive materials (environmental factor) are the limiting factors for the amounts of FC which can be taken up in cement clinker production. However, FC uptake contributes to reduce the amount of landfilled BR in aluminium production. The ecotoxicological study indicates that FC and BR can be grouped into a similar ecotoxicity category: slightly ecotoxic. Therefore, there is no additional risk to the environment by transforming the original waste (BR) into another type (FC). Thus, landfilling same amounts of BR and FC results in similar environmental impacts. However, only the amount of FC exceeding the demand of the cement industry located in the proximity of the aluminium plant might be landfilled.

The more environmentally friendly production of ScF_3 by antisolvent crystallisation and calcination compared to gas fluorination enables the direct production of AlSc alloys by an electrochemical process, which is not further discussed in this work.

The GWP value of Sc_2O_3 produced with the SCALE production route is about half the GWP value for the state-of-the-art production of Sc_2O_3 from REE tailings when applying equal allocation principles as used in the LCA of Wang et al. (2020).

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

The data that has been used is confidential.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.clwas.2024.100129.

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