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Life cycle assessment of zinc and iron recovery from spent pickling acids by membrane-based solvent extraction and electrowinning

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ABSTRACT

In this paper we conducted a life cycle assessment to evaluate the environmental performance of the valorization of spent pickling acid (SPA) generated in the hot-dip galvanizing (HDG) process. We analyzed the environmental impacts of treating one m^3 of SPA, comparing the reference treatment consisting of neutralization, precipitation, stabilization, and landfilling of the metallic sludge (scenario #1), with the innovative LIFE2ACID technology (scenario #2) that produces secondary zinc and iron chloride in solution through non-dispersive solvent extraction (NDSX) and electrowinning (EW). The results showed that the materials credits achieved by the implementation of LIFE2ACID technology turned most of the impact categories evaluated (toxicity, acidification, eutrophication, ozone depletion, etc.) into environmental benefits. Scenario #2 was adapted to achieve either zinc-only recovery (#2.1) or simultaneous iron and zinc recovery (#2.2). The abiotic depletion potential (ADP) of fossil fuels increased slightly from scenario #1 to scenario #2.1 because of the higher energy demand and NaOH consumption of EW, and because only zinc was recovered. However, the valorization of both zinc and iron chloride in scenario #2.2 reduced the ADP-fossil by 27%, compared to the reference treatment. Furthermore, the global warming impact was reduced by 20% and 97% in scenarios #2.1 and #2.2, respectively. With the focus on promoting the circular economy concept, we conclude that the LIFE2ACID technology significantly improves the environmental performance of SPA management. Next steps should consider the life-cycle costs analysis in specific scenarios to find out the trade-off between environmental and economic objectives.

1. Introduction

The increasing pressure on natural resources, as well as the loss of valuable materials, have led to strengthen the focus on metals recovery from waste streams (Allegrini et al., 2015). Thus, strategies that promote the circularity of metal cycles are rapidly gaining interest. Metal recycling reduces both the use of virgin materials (Santero and Hendry, 2016) and the energy demand in primary materials production (Raghupathy and Chaturvedi, 2013). Nonetheless, the consequences of replacing primary materials are not straightforward, because increasing the quantity of recycled materials in each given product has a much broader impact resulting from alterations in the other systems that are all linked through the resource flows (Chang et al., 2019).

Zinc is the fourth most used metal after iron, aluminum and copper (Guo et al., 2010). However, zinc ore mines are finite and their reserves are insufficient to supply the predicted demand (Ng et al., 2016). Indeed, a recent study has estimated that cumulative primary production of zinc will exceed reserves in 2025 (Watari et al., 2021). Currently, around 25–30% of global zinc demand is supplied from recycled zinc, as reported by Kaya et al. (2020). In this scenario, metal zinc mining from the spent pickling acid (SPA) of hot-dip galvanizing (HDG) becomes an attractive option for metal waste recyclers and zinc users (Zueva et al., 2021). SPA is the main liquid waste of HDG industrial facilities, in which average concentrations of 101.6 and 95.7 g L^{-1} of zinc and iron have been reported (Arguillarena et al., 2020a), together with low and trace amounts of manganese, nickel, lead and copper. The reference treatment of SPA consists of neutralization/precipitation (N/P) using an alkaline agent (Diban et al., 2011), followed by solidification/stabilization (S/S) by means of binder agents (Conner and Hoeffner, 1998) and landfilling. Nevertheless, the practice of industrial landfilling creates several environmental issues such as the loss of valuable resources, the lack of space available for disposal of hazardous residues and the long-term lixiviation of heavy metals (Devi et al., 2014). Therefore, strategies designed to reduce waste generation and increase resource efficiency are essential for improving the environmental and economic performance of zinc coating industrial activities (García et al., 2013). In parallel, the recovery of metal resources can reduce the environmental burdens of the

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Nomenclature

ABBREVIATIONS

ADP-elements	Abiotic Depletion of Elements
ADP-fossil	Abiotic Depletion of Fossil Fuels
AP	Acidification Potential
CED	Cumulative Energy Demand
EP	Eutrophication Potential
EW	Electrowinning
FAETP	Freshwater Aquatic Ecotoxicity Potential
GTP	Global Toxicity Potential
GWP	Global Warming Potential
HDG	Hot-dip galvanizing
HTP	Human Toxicity Potential
ILCD	International Reference Life Cycle Data System
LCA	Life cycle assessment
LCI	Life cycle inventory
LCT	Life cycle thinking
MAETP	Marine Aquatic Ecotoxicity Potential
N/P	Neutralization/Precipitation
NDSX	Non-dispersive solvent extraction
ODP	Ozone Depletion Potential
PAH	Polycyclic aromatic hydrocarbon
POCP	Photochemical Ozone Creation Potential
S/S	Solidification/Stabilization
SHG	Special high-grade
SPA	Spent pickling acid
TETP	Terrestrial Ecotoxicity Potential

metal mining industry (García et al., 2014). In this regard, a recent study reported a hydrometallurgical process for the recovery of zinc in the form of high-grade zinc salts from the dross waste produced by galvanizing (Sinha et al., 2020). Similarly, the sludge resulting from steel manufacturing blast furnace smoke has been used for the selective recovery of zinc and manganese (Mocellin et al., 2015).

Applying a new perspective, the LIFE2ACID project approaches the recovery of iron and zinc from SPA by a combination of non-dispersive solvent extraction (NDSX) and electrowinning (EW) technologies. The former employs hollow-fiber membrane contactors to perform the solvent extraction and back-extraction of metals (Urtiaga et al., 2010) making it possible to separate and purify the zinc contained in SPA. Electrowinning, as has been previously demonstrated at laboratory scale, allows zinc electrodeposition of purified zinc solutions (Carrillo-Abad et al., 2015). The remaining SPA is recovered as a ferrous chloride ($FeCl_2$) solution, which has applications in wastewater treatment plants as hydrogen sulfide suppressor of the biogas produced in anaerobic digesters (Kulandaivelu et al., 2020). The NDSX/EW technology has already been demonstrated at a pilot plant scale within the scope of the LIFE2ACID project (Arguillarena et al., 2020a) using previous experience gained at the laboratory scale (Laso et al., 2015). Nevertheless, knowing the environmental burdens of NDSX/EW is a key factor in comparing the innovative LIFE2ACID technology with the reference SPA treatment (N/P, S/S, and landfill), which has not been analyzed yet.

Life cycle assessment (LCA) must play a role in supporting the appraisal of metal recovery technologies. Indeed, several LCA studies have already assessed the environmental impacts of metal recovery from different residues. Rajaeifar et al. (2021) investigated the recycling of spent lithium-ion batteries by means of pyrometallurgical processes in terms of global warming potential (GWP) and cumulative energy demand (CED). Bigum et al. (2012) conducted the LCA of the recovery of aluminum, copper, gold, iron, nickel, palladium and silver from high-grade waste electrical and electronic equipment using a metallurgic treatment. The recovery of these metals showed significant savings concerning environmental loads and resource consumption, although these credits were underestimated due to the lack of adequate data from mining and refining of ores used to model the avoided burdens of the recovered metals. Amann et al. (2018) compared the environmental impacts of 18 phosphorous recovery technologies from municipal wastewater in terms of CED, GWP and acidification potential (AP). Some of these technologies had a higher environmental impact than the reference system, which was the wastewater treatment plant without phosphorous recovery, so their future implementation should be avoided unless further innovation improves their environmental performance. However, the analysis of a case of study closer to the herein presented was carried out by García et al. (2013), where the authors analyzed the implementation of the eco-innovative emulsion pertraction technology for the selective separation of zinc and iron impurities from spent chromium conversion baths formulated with trivalent chromium. Overall, the literature analysis led us to conclude that there is still a lack of LCA studies on technologies for recovering metals from the spent baths generated during the coating of metal surfaces. Life cycle thinking (LCT) should become a key requirement to ensure that we choose the most appropriate management alternative for SPA, considering all aspects of the system under study over its entire life cycle. This means that the study includes all the stages from the extraction of raw materials and the energy generation to waste treatment itself and the recovery or final disposal.

This work is aimed at the LCA of the treatment of spent pickling acids of hot dip galvanizing. The technical alternatives that are analyzed and compared are the reference treatment that applies N/P, S/S and landfilling, and the innovative NDSX/EW technology, which allows the recovery of zinc and iron metal resources. The function of the reference technology is to treat SPA waste; however, the NDSX/EW provides this one and the additional function of metals recovery. To compare these alternatives, it is required that both of them provide the same functions. So, it was necessary to identify and calculate the materials credits of the recovered metallic resources, iron and zinc, which substitute primary materials. In addition, the life cycle inventory (LCI) was built using high quality data that were collected in an HDG industrial plant and during the large-scale treatment of real SPA in a NDSX/EW pilot plant. The comparison and alternatives selection were based on the indicators used in the CML method, aim at quantifying the consumption of resources, the effect on climate change and the toxicity impaired on air, water and soil. Overall, this study contributes to filling the gap of studies aimed at the environmental analysis of SPAs treatment and the surface treatment sector.

2. Methodology

In the present study we applied the LCA methodology according to the principles and requirements of ISO 14040 (2006) and ISO 14044 (2006). We used Gabi 9.2 software and the Ecoinvent and professional Sphera databases.

2.1. Goal and scope definition

The goal of this LCA was to assess the resource usage and environmental impacts of the management of SPA by comparing the reference treatment with the innovative NDSX/EW technology. The LCA of a waste or wastewater management system is divided in the same stages that the LCA of a product. The main difference between both LCAs resides in what it is meant by cradle and grave. Whilst it shares the same grave as individual products, the lifecycle of waste does not share the same cradle (Margallo, 2014). In this work, the process starts at the end-of-life of the SPA and ends at the obtention of secondary material resources. Therefore, the study considers a 'grave to cradle' perspective, which includes the SPA treatment process itself, the extraction and production of raw materials required, and the management of the metal sludge generated. The reference treatment (N/P, S/S, and landfilling) and the NDSX/EW technology will be referred to as scenarios #1 and #2, respectively, throughout this study. Furthermore, scenario #2 can be adapted in order to pursue two different objectives: the recovery of zinc only (scenario #2.1) or both iron and zinc recovery (scenario #2.2). In this study, the recovered zinc replaced primary zinc and the recovered iron replaced commercial iron (III) chloride solutions (40 wt% $FeCl_3$). Fig. 1 shows the system boundaries of scenarios #1, #2.1 and #2.2.

The main function was to treat the SPA produced during HDG to avoid the release of heavy metals into the environment, meet the wastewater discharge standards set by the Industrial Emissions Directive (European Council, 2010) and comply with the heavy metal leaching limits (European Council, 2003). Consequently, the functional unit in both scenarios was one m^3 of SPA. The use of one m^3 as a functional unit is very common in wastewater treatment systems, as previously reported in similar works (Dominguez et al., 2018). Nevertheless, scenarios #2.1 and #2.2 incorporated additional functions involving substitution of primary zinc and commercial $FeCl_3$.

In scenario #1, calcium hydroxide ($Ca(OH)_2$) precipitated the metal hydroxides, and the resulting hazardous metal sludge should be sent to a hazardous waste landfill site. However, the subsequent S/S treatment of the sludge using Portland cement would allow its disposal in a non-hazardous waste landfill site.

In scenario #2, zinc was selectively extracted in hollow fiber modules with total membrane area of 80 m^2 that facilitated the non-dispersive contact of the SPA and the organic extractant phase (Alonso et al., 1997). The extractant was regenerated with water (Ortiz et al., 2004), allowing the transfer of zinc to the aqueous stripping, and it was then reused in subsequent extraction/stripping cycles (Arguillarena et al., 2020a). The main inputs of the NDSX were electricity to pump the fluid phases and tap water to strip and wash the organic phase.

The stripping phase from the NDSX was mostly a zinc chloride ($ZnCl_2$) aqueous solution that was introduced in the EW stage, where it required pretreatment (adjust the pH to 3–4, aeration, flocculation, precipitation and filtration) to remove traces of iron that would otherwise negatively affect the quality of the zinc deposit. Next, the electrodeposition of zinc took place on the cathodes in the electrochemical reactor, using the electricity supply as the main input, whereas, sodium hydroxide (NaOH) was added to maintain the EW unit at the appropriate pH. The clarified water obtained after zinc electrodeposition was returned to the NDSX stage to be reused as stripping water. The stripping phase also had a high chloride content which was oxidized on the anode to give chlorine gas. Chlorine can be used to produce sodium hypochlorite (NaClO), thereby implying the consumption of NaOH. Finally, titanium plates coated with mixed metal oxides were used as anodes in the EW reactor. Zinc plate cathodes were reused in several batches to extend their lifespan and then were smelted together with the main zinc deposit.

Finally, the yield of zinc extraction and the composition of the SPA can be varied by selecting appropriate NDSX operation conditions. In scenario #2.1, the remaining SPA contains most of the initial iron but still contains some zinc, so this stream requires additional treatment. In scenario #2.2, the intensification of the extraction process achieved very high zinc extraction rates, converting the SPA into an iron (II) chloride solution that could be used in wastewater treatment systems. Nevertheless, achieving the full recovery of $FeCl_2$ and $ZnCl_2$ in scenario #2.2 required operation changes that resulted in higher energy and water consumption compared to scenario #2.1.

2.1.1. Cut-off criteria

This work establishes that each excluded material flow must not exceed 1% of the mass of each unit process while the sum of all excluded material flows in the system must not exceed 5% of the total mass flux. We considered all the energy flows in the system. However, the production of the organic phase was not included in the system boundaries because it was continuously regenerated and reused within the system meaning that losses were negligible. The residual SPA was not considered as it represented less than 1% of the mass in the NDSX process (0.001–0.002 m^3 residual SPA/m^3treated SPA). Finally, the production of titanium anodes was not included within the system boundaries because, in addition to their extended lifespan (≃10 years), they were also reused. Zinc cathodes were also not included in the system boundaries because they were reused.

2.1.2. Allocation

The main function in scenarios #1, #2.1 and #2.2 was the management of the SPA waste generated by the HDG process. Nevertheless, scenarios #2.1 and #2.2 were defined as multi-output processes that provided an additional function to the system: scenario #2.1 included the production of primary zinc and scenario #2.2 the obtention of

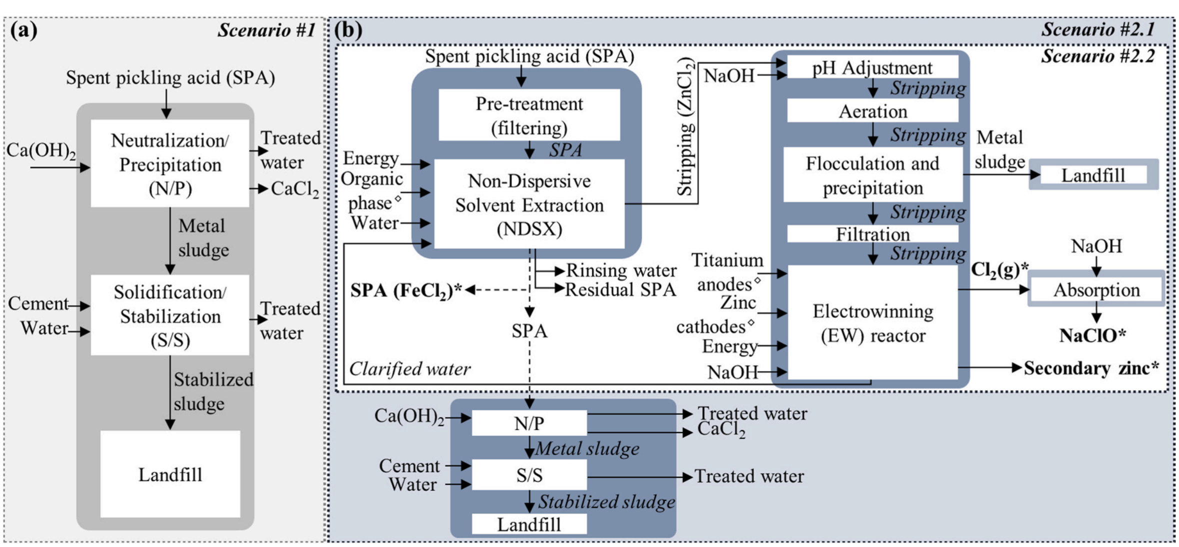

Fig. 1. System boundaries of (a) scenario #1 in the block with light grey background: neutralization/precipitation (N/P), solidification/stabilization (S/S) and landfilling, and (b) scenarios #2.1 (white background) and #2.2 (light blue background): non-dispersive solvent extraction and electrowinning (NDSX/EW) technology (*recovered products). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Life cycle inventory of scenario #1 per functional unit (one m^3 of SPA).

Inputs	Units	Value
SPA	m^3	1.0
$Ca(OH)_2$	kg/m^3 SPA	396.8
Portland cement		22.4
Tap water		52.2
Outputs	**Units**	**Value**
Treated water (after N/P)	kg/m^3 SPA	97.6
$CaCl_2$ (after N/P)		594.4
Treated water (after S/S)		869.1
Stabilized sludge		273.4

primary zinc and commercial iron chloride. Based on the ISO 14040 (2006) guidelines, the environmental burdens in such multifunctional systems must be divided into different functions through system division or expansion, avoiding the use of causality and non-causality allocation. System expansion means that the boundaries of the system investigated are expanded to include the alternative production of exported functions (Ekvall and Finnveden, 2001). The calculation of recycling credits is one of the main challenges in end-of-life modelling (Koffler and Finkbeiner, 2018). Zinc from the electrowinning substitutes primary zinc that is mainly used in steel galvanization to increase the durability and lifetime of steel products (Arguillarena et al., 2020b). Iron as $FeCl_2$ substitutes commercial $FeCl_3$ that is employed for hydrogen sulfide removal in the anaerobic sludge digesters at wastewater treatment plants. Previous research analyzing the substitution of $FeCl_3$ for $FeCl_2$ has shown excellent results (Kulandaivelu et al., 2020). The same procedure was applied to include the NaClO credits in scenarios #2.1 and #2.2., as explained in detail in section 3.5.

2.2. Life-cycle inventory

Tables 1 and 2 show the LCI for scenarios #1, #2.1 and #2.2 while section 1 of the supplementary material details the calculations of the inventory inputs and outputs. The LCI of scenarios #2.1 and #2.2 were obtained using semi-empirical models that had been previously validated and adjusted according to the results of pilot-scale demonstration experiments (Arguillarena et al., 2020a). In experiments aimed at zinc recovery (scenario #2.1), the SPA treatment capacity of the NDSX/EW pilot plant was 26.6 m^3 SPA/year. When both iron and zinc were recovered, more time was needed to achieve the required separation and the capacity of the pilot plant was reduced to 8 m^3 SPA/year. The unit processes used to model the scenarios from the Ecoinvent and professional Sphera databases are collected in Table S2 (supplementary material). The electricity consumed by the NDSX/EW technology is based on the Spanish electricity grid mix, which is similar to the European grid mix. This grid mix is mainly composed by natural gas (21.4%), nuclear (20.3%), wind (18.6%), hard coal (13.0%) and hydro (13.4%). Other sources are fuel oil, photovoltaic, biomass, solar thermal, lignite, coal gases, waste and biogas.

2.3. Life-cycle impact assessment

We selected the midpoint CML 2001 method as the impact assessment method. Corominas et al. (2020) reviewed 121 papers in the wastewater treatment sector and reported that 36% of total studies used CML, and the rest of the studies were distributed among ReCiPe, TRACI and the International Reference Life Cycle Data System (ILCD) Handbook users. Regarding the selection of key impact indicators, climate change, eutrophication and ecotoxicity are recommended for the sustainability analysis of wastewater systems (Corominas et al., 2020). Other studies that evaluated the impact of the production of galvanized steel in Spanish industrial facilities (Arguillarena et al., 2020b), as well as the sectoral Environmental Product Declaration at European level (EGGA, 2016) and the environmental studies of hot-dip galvanized steel carried out by the American Galvanizers Association (AGA and IZA, 2017), analyzed the 11 categories of the CML. Therefore, this work includes the following CML impact categories: Abiotic Depletion of Elements (ADP-elements) [kg Sb eq.], Abiotic Depletion of Fossil Fuels (ADP-fossil) [MJ], Acidification Potential (AP) [kg SO_2 eq.], Eutrophication Potential (EP) [kg PO_4^{3-} eq.], Freshwater Aquatic Ecotoxicity Potential (FAETP) [kg DCB eq.], Global Warming Potential (GWP) [kg CO_2 eq.], Human Toxicity Potential (HTP) [kg DCB eq.], Marine Aquatic Ecotoxicity Potential (MAETP) [kg DCB eq.], Ozone Layer Depletion Potential (ODP) [kg R11 eq.], Photochemical Ozone Creation Potential (POCP) [kg ethene eq.] and Terrestrial Ecotoxicity Potential (TETP) [kg DCB eq.].

3. Results and discussion

3.1. Environmental impacts of scenario #1: reference treatment of spent pickling acids

In this section (Fig. 2) we analyze the environmental impacts of the reference SPA treatment (scenario #1). The N/P stage is represented by

Table 2
Life cycle inventory of scenarios #2.1 and #2.2 per functional unit (one m^3 of SPA).

		Non-dispersive solvent extraction (NDSX)		Electrowinning (EW)	
Inputs	Units	Scenario #2.1	Scenario #2.2	Scenario #2.1	Scenario #2.2
SPA	m^3	1.0	1.0	–	–
$Ca(OH)_2$ (N/P)	kg/m^3 SPA	306.6	–	–	–
Portland cement (S/S)		16.4	–	–	–
Tap water (S/S)		38.2	–	–	–
Electricity	MJ/m^3 SPA	71.2	235.5	670.0	1,587.6
Tap water for stripping	kg/m^3 SPA	620.4	1,470.0	–	–
100% wt. NaOH (pre-treatment EW)		–	–	18.9	44.8
100% wt. NaOH (EW reactor)		–	–	7.6	17.9
100% wt. NaOH (Cl_2 absorption)		–	–	46.5	110.2
Outputs	Units	Scenario #2.1	Scenario #2.2	Scenario #2.1	Scenario #2.2
Rinsing water (after NDSX)	kg/m^3 SPA	620.4	1,470.0	–	–
Treated water (after N/P)		78.0	–	–	–
$CaCl_2$ (after N/P)		459.6	–	–	–
Treated water (after S/S)		986.9	–	–	–
Stabilized sludge (after S/S)		199.8	–	–	–
Metal sludge (pre-treatment EW)		–	–	16.1	17.6
$FeCl_3$ without water (40 wt% $FeCl_3$)		–	265.2	–	–
Secondary zinc		–	–	53.3	78.9
NaClO (175 g Cl_2/L)		–	–	407.0	602.8

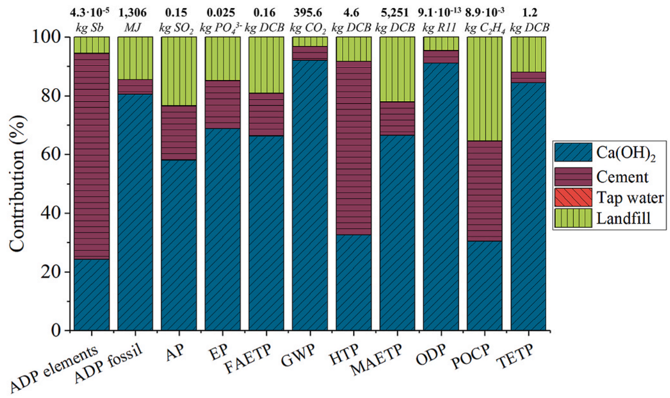

Fig. 2. Relative contributions of reference SPA treatment (scenario #1) to the total impact (burden) of N/P, S/S and landfilling. The total value of each impact category is provided on the top of each bar. Functional unit: one m^3 of SPA treated.

the production of $Ca(OH)_2$ and the S/S by the consumption of cement and tap water. The absolute impact values are collected in Table S3 in the supplementary material. The main contributors in all the impact categories were the stages of N/P and S/S because of the burdens of $Ca(OH)_2$ and cement production. Landfilling also contributed to all the indicators, but to a lesser extent. The influence of the water consumption for S/S was lower than 1% of the total impact in all the categories, therefore it was negligible compared to the other contributions. Cement production was the main contributor to ADP-elements and HTP. The ADP-elements indicator is derived from the consumption of gypsum, while the HTP element comes from the emission of polycyclic aromatic hydrocarbons (PAHs) into the air. The release of PAHs depends on the type of fuel, raw material and operational conditions of the combustion.

For the ADP-fossil, AP, EP, FAETP, GWP, MAETP, ODP and TETP categories the impact was mainly due to the production of $Ca(OH)_2$ that is consumed in the N/P stage. For the ADP-fossil category the requirements of lignite, natural gas, and hard coal in the thermal decomposition of calcium carbonate/limestone into quicklime (CaO), which is needed to produce $Ca(OH)_2$, demanded a lot of fossil fuel resources (Kumar et al., 2007). The impact categories AP and EP were driven by the emissions of nitrogen oxides (NO_x) and sulfur dioxide (SO_2) resulting from the energy transformation needed for limestone calcination (OEDC, 2013). In the case of FAETP and TETP, both indicators were increased by the emission of heavy metals to freshwater and air, especially vanadium and nickel in FAETP, and mercury in TETP. The CO_2 emissions produced during the combustion of fossil fuels to fulfil the energy requirements of $Ca(OH)_2$ fabrication justified GWP. The category MAETP was the result of the emission of hydrogen fluoride (HF) into the air, while ODP was totally due to the emissions of chloromethane into the air. The causes of POCP were shared between $Ca(OH)_2$ production, the use of cement and landfilling.

Overall, the production of $Ca(OH)_2$ was the main cause of the environmental impacts in scenario #1 because of the higher consumption of $Ca(OH)_2$ in the N/P stage (396.8 kg $Ca(OH)_2/m^3$ SPA) with respect to the amount of cement used in S/S (22.4 kg cement/m^3 SPA). Nonetheless, the S/S was carried out to stabilize the metallic sludge so that it could be disposed of in a non-hazardous waste landfill site.

3.2. Environmental impacts of scenario #2.1: zinc recovery with non-dispersive solvent extraction and electrowinning

Section 3.2 evaluates the environmental burdens of the NDSX/EW technology in scenario #2.1. The contribution of raw materials production, energy generation and waste management to the environmental burdens of scenario #2.1 are shown in Fig. 3. The material credits gained by recovering zinc have been included. Complementary,

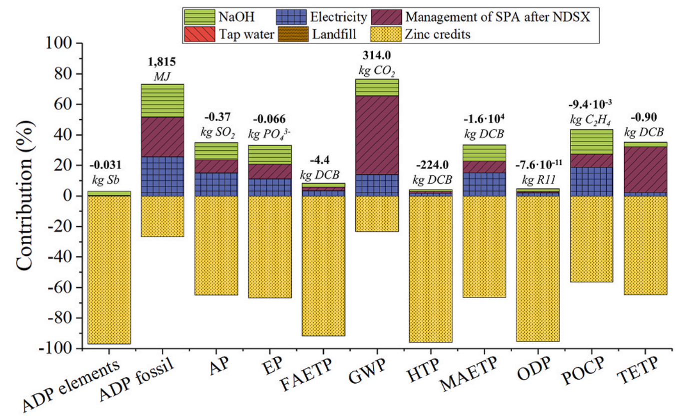

Fig. 3. Relative contributions to the total impact of zinc recovery with non-dispersive solvent extraction and electrowinning (NDSX/EW) in scenario #2.1 including the material credits gained in the process. The total value of each impact category is given on the top of each bar. Functional unit: one m^3 of SPA treated.

Table S4 in the supplementary material presents the absolute impacts of scenario #2.1. The NDSX stage consumed electricity for fluid pumping, water as a stripping agent, and the SPA remaining after NDSX required management through the reference treatment. EW also required the use of electricity for zinc electrodeposition, NaOH consumption, and the landfilling of the metal sludge generated in the pre-treatment.

In scenario #2.1, the EW reactor accounted for most of the electricity consumption. The impact of tap water production and consumption and the landfilling of the metal sludge generated in the EW pre-treatment was negligible compared to the consumption of electricity and NaOH. Furthermore, all the impact categories except ADP-fossil and GWP had negative values as a result of the burdens avoided through the substitution of primary zinc by the secondary zinc obtained using the NDSX/EW technology. Hence, in most impact categories except for ADP-fossil and GWP, the environmental benefits of zinc recovery and recycling exceeded the environmental impacts of NDSX/EW. However, these benefits were influenced by the characterization factors of special high-grade (SHG) zinc, which were much higher than those of the zinc produced by the other processes. These differences explain the greater contribution of zinc to the material credits in scenario #2.1 because of the environmental burdens it avoids.

The contribution and causes of the most relevant production process (namely electricity generation, NaOH production, SPA management after NDSX and zinc credits) to the impact categories are explained below. None of the processes involved in the LIFE2ACID technology strongly contributed to the ADP-fossil category, although the use of non-renewable resources like natural gas and hard coal stood out. The zinc credits obtained through this process allowed the reduction of the ADP-fossil impact category because of the avoided consumption of hard coal and natural gas in the processing of ores to extract and refine primary zinc. The category GWP was mainly increased by the CO_2 emissions from the production of the $Ca(OH)_2$ used in treatment of the residual SPA after NDSX treatment. Regarding zinc credits, GWP is the least influenced category by the environmental benefits of zinc recovery.

The zinc credits obtained in this process considerably reduced the ADP-elements factor because less zinc mineral was used. Moreover, FAETP and HTP were decreased because the emission of heavy metals into freshwater was prevented, while the ODP reduction was entirely due to the avoided chloromethane emissions into the air. The avoided emissions of PAH into air allowed the further reduction of HTP. The categories AP, EP, MAETP, POCP and TETP were also strongly influenced by the award of zinc credits (although to a lesser extent) because of the avoided NO_x, SO_2 and heavy metal emissions into the air. Finally, after considering the zinc credits, the production of $Ca(OH)_2$ and NaOH,

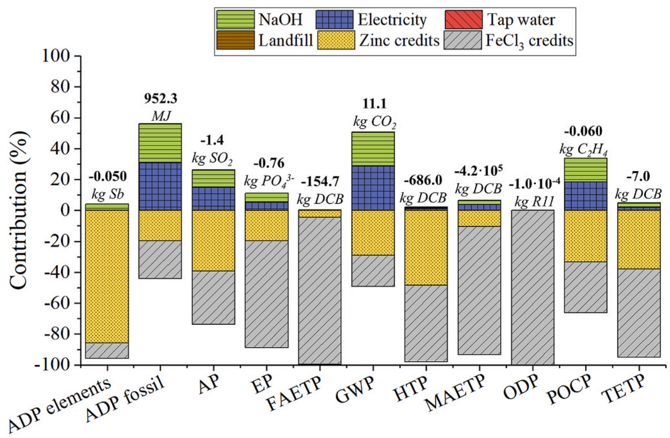

Fig. 4. Relative contributions to the total impact of non-dispersive solvent extraction and electrowinning (NDSX/EW) zinc and iron recovery in scenario #2.2 while considering material credits. The total value of each impact category is on the top of each bar. Functional unit: one m^3 of SPA treated.

as well as the electricity consumption by EW, justified most of the impact category contributions.

3.3. Environmental impacts of scenario #2.2: zinc and iron recovery with non-dispersive solvent extraction and electrowinning

This section analyzed the environmental impacts of the NDSX/EW technology when applied to recover both zinc and iron (scenario #2.2 in Fig. 1). Fig. 4 shows the results of the percentage contribution of the main processes to the impact categories while the details of the absolute magnitudes are presented in Table S5 (supplementary material).

Most impact categories in scenario #2.2 were strongly influenced by

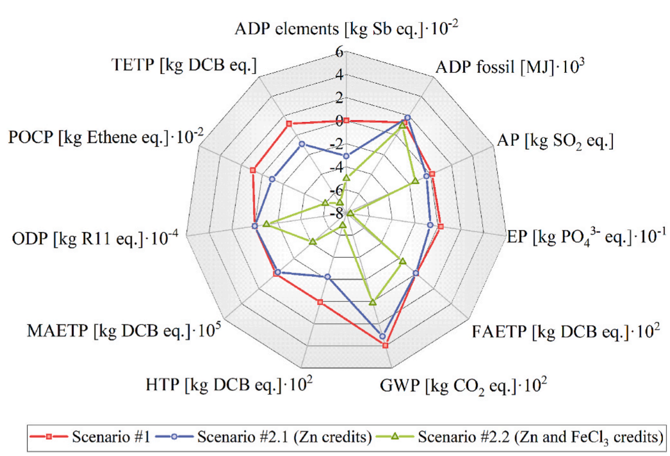

Fig. 5. Environmental impacts of (■) scenario #1: neutralization/precipitation (N/P), solidification/stabilization (S/S) and landfilling, (●) scenario #2.1: NDSX/EW with zinc recovery (including zinc credits) and (▲) scenario #2.2: NDSX/EW with zinc and iron recovery (including zinc and FeCl$_3$ credits). Functional unit: one m^3 of SPA treated.

the zinc and iron credits, except for ADP-fossil and GWP. The global value of ADP-fossil and GWP was shared similarly between the increasing impact of both the electricity generation and NaOH production and the impact reduction assigned to the material credits. The further consideration of zinc credits, as already analyzed in section 3.2, caused an additional reduction in the ADP-elements indicator because of the avoidance of the burdens derived from the use of zinc mineral. In a similar way to the results already presented in section 3.2, the material credits outweighed the environmental impacts of electricity generation and production of NaOH and $Ca(OH)_2$ because of the higher characterization factors of primary zinc and $FeCl_3$. Nonetheless, the material credits for scenario #2.2 were more intensely influenced by the higher iron chloride recovery compared to zinc recovery (265.2 kg $FeCl_2$/m^3 SPA vs. 78.9 kg secondary zinc/m^3 SPA).

3.4. Comparison of SPA treatment approaches: reference treatment versus NDSX and EW as metal recovery technologies

In this section, we compare the environmental impacts of scenarios #1, #2.1 and #2.2 considering the material credits obtained for zinc and iron chloride (Fig. 5). Overall, scenario #2.2 was the most environmentally friendly option because of the higher recovery of zinc and iron chloride. Every impact category reached its minimum value in scenario #2.2, although there were significant differences in the percentage reductions.

Next, we will focus on analyzing the impact categories related to energy consumption and toxicity. We will not discuss the ADP-elements impact category in detail because NDSX/EW allows the recovery of zinc and iron (II) chloride, so the abiotic resource savings are evident. Table 3 compares the ADP-fossil and GWP indicators, reflecting the impact of the energy consumption of NDSX/EW. However, ADP-fossil and GWP did not follow the same trend when moving from the reference treatment to the NDSX/EW alternative with zinc recovery or with zinc and iron recovery. In this sense, GWP decreased by 21% and 97% in scenarios #2.1 and #2.2, respectively, compared to the reference treatment represented in scenario #1. The higher GWP of scenario #1 was due to the CO_2 emissions into the air caused by $Ca(OH)_2$ production as an essential input of the N/P stage. However, there was an increase in the ADP-fossil factor in scenario #2.1, revealing that the energy consumption of zinc electrodeposition had offset the benefit of the zinc material credits and that the energy consumption of EW should be minimized to avoid the abiotic depletion of fossil fuels. The recovery of zinc and iron in scenario #2.2 resulted in the lowest impact for ADP-fossil from among the scenarios we studied in this current work.

Furthermore, the indicators related to the toxicity impacts on human health and ecosystems can be gathered into a global toxicity indicator. Thus, we defined a global toxicity potential (GTP) indicator by summing the individual values of the FAETP, MAETP, HTP and TETP (which all share the same units in CML 2001), as shown in Table 4.

The reference treatment (scenario #1) presented the highest toxicity with a GTP of 5,257 kg DCB eq./m^3 SPA. Certainly, the material credits of zinc and iron (II) chloride strongly reduced this relative toxicity. The GTP index presented negative values in scenarios #2.1 and #2.2, indicating that the substitution of primary materials by secondary materials reduced the toxicity stress associated with the extraction of minerals and metallurgical processes. The impact category FAETP was reduced tremendously when iron (II) chloride credits were considered in

Table 3
Indicators related to energy consumption: ADP-fossil and GWP. Comparison of the SPA reference treatment (scenario #1) and the NDSX/EW technology in scenarios #2.1 (zinc recovery) and #2.2 (zinc and $FeCl_2$ recovery).

Impact category	Units	Scenario #1	Scenario #2.1	Scenario #2.2
ADP-fossil	MJ/m^3 SPA	1,306	1,815	952.3
GWP	kg CO_2 eq./m^3 SPA	395.6	314.0	11.1

Table 4
Toxicity indicators for the SPA reference treatment (scenario #1) and the NDSX/EW technology in scenarios #2.1 (zinc recovery) and #2.2 (zinc and $FeCl_2$ recovery).

Toxicity	Impact category	Units	Scenario #1	Scenario #2.1	Scenario #2.2
Freshwater	FAETP		0.16	−4.4	−154.7
Sea water	MAETP		5,251	$-1.7\cdot10^4$	$-4.2\cdot10^5$
Human health	HTP	kg DCB eq./m^3 SPA	4.5	−224.0	−686.0
Terrestrial	TETP		1.2	−0.90	−7.0
Global Toxicity	GTP		**5,257**	$-1.7\cdot10^4$	$-4.2\cdot10^5$

scenario #2.2 because of the avoided emissions of heavy metals into freshwater. The highest contribution to the toxicity index compared with the other indicators was MAETP, which was caused by the emission of HF into the air. Finally, HTP showed the same trend as MAETP, but to a lesser extent. Of note, the MAETP category has been the subject of some academic discussion (Eljaddi et al., 2021) because of the characterization factor ($4.1\cdot10^7$ kg DCB eq./kg HF) used to calculate HF emissions in the CML impact assessment method. This characterization factor may be too high, consequently resulting in an overestimation of the potential environmental impact of HF emissions (Koornneef et al., 2008). However, even if they were overrated, the HF emissions attached to fossil fuel combustion should not be neglected (Ligthart et al., 2010). The CML characterization factors for HF are based on considering the long residence times of HF in the air (van Harmelen et al., 2007). Nevertheless, considering the elevated HF solubility in water at normal ambient temperatures, it is believed that the monomer and oligomers of HF will dissociate once they are in the aqueous phase (Cheng, 2018). Therefore, the HF characterization factors applied in the literature should be decreased by a factor of about 80 to account for the incorrect assumption that HF remains in the air for long periods.

3.5. Material credits of sodium hypochlorite after zinc electrowinning

In this section, we evaluate the additional material credits gained through the recovery of sodium hypochlorite in the EW reactor. In the NDSX step of the SPA treatment, both chloride and protons accompany zinc mass transfer (Arguillarena et al., 2020a). Next, the electrochemical reduction of zinc cations occurs on the cathode in the EW reactor, while chloride is oxidized on the anode to form chlorine (Eq. 5 and Eq. 6 in the supplementary material). Chlorine gas is a powerful oxidant which is very irritating and corrosive (Evans, 2015). Among the different reaction systems used to remove chlorine from industrial gas streams, absorption in a sodium hydroxide solution is the most extended one (Agrawal, 2013). Therefore, we treated the electrogenerated chlorine gas with NaOH, producing a NaClO solution, as shown in Fig. 1. Obtaining a new product (NaClO) implies the consumption of NaOH, whose production process is very intensive in terms of the use of resources. Fig. 6 shows the environmental impacts of scenarios #2.1 and #2.2, either with or without the NaClO credits. The absolute values of scenarios #2.1 and #2.2 with NaClO credits in addition to zinc and iron (II) chloride credits are collected in Table S6 in the supplementary material.

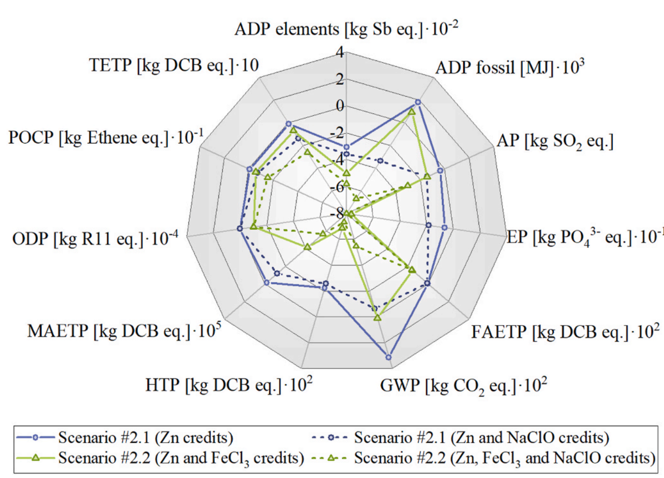

Fig. 6. Environmental impacts of (—○—) scenario #2.1: non-dispersive solvent extraction and electrowinning (NDSX/EW) with zinc credits, (--○--) scenario #2.1: NDSX/EW with zinc and NaClO credits, (—△—) scenario #2.2: NDSX/EW with zinc and $FeCl_3$ credits and (--△--) scenario #2.2: NDSX/EW with zinc, $FeCl_3$ and NaClO credits.

Scenario #2.2 with zinc, $FeCl_3$ and NaClO credits resulted in the lowest environmental impact compared with the other options because it maximized the recovery of products per m^3 of SPA. Scenario #2.1 recovered 53.3 kg Zn/m^3 SPA and 407 kg NaClO/m^3 SPA. In contrast, scenario #2.2 recovered 78.9 kg Zn/m^3 SPA, 265.2 kg iron chloride/m^3 SPA and 602.8 kg NaClO/m^3 SPA. Thus, considering the characterization factors per kilogram of SHG zinc, commercial $FeCl_3$ and NaClO (175 g Cl_2/L), zinc credits could provide the maximum environmental benefits. Nevertheless, as NaClO was recovered in larger quantities than the other products, its material credits strongly influenced the total environmental impact and therefore, the environmental impacts of scenarios #2.1 and #2.2 were considerably reduced when NaClO was recovered. The environmental benefits of NaClO recovery in scenario #2.2 were notable compared to the other scenarios using NDSX/EW technology, even though its recovery involves the consumption of NaOH in the absorption process.

4. Conclusions

This work studied the environmental impacts and the raw materials usage of the recovery of zinc and iron chloride from the SPA, which is one of the most voluminous wastes of HDG, through the use of NDSX combined with EW. We compared this innovative technology with the reference treatment consisting of N/P, S/S and landfilling. Our findings identified the main environmental advantages of NDSX/EW, thereby contributing to the current academic literature related to this field.

The environmental impacts of the reference SPA treatment were higher than those of the NDSX/EW technology in all the scenarios adapted either to the goal of recovering only zinc or for the simultaneous recovery of zinc and iron chloride. Although NDSX/EW implies a higher consumption of materials and energy, the main advantage of this aforementioned technology came from the material savings of the recovered metal products: zinc and $FeCl_2$, which can substitute primary zinc and commercial $FeCl_3$. When comparing the impacts of the reference treatment and the NDSX/EW technology, GWP was reduced by more than 97% when iron and zinc were recovered simultaneously. The same trend occurred for the ADP-fossil indicator, which was reduced by 27%. The ADP-elements indicator was also notably improved because primary zinc production is very resource-intensive. Furthermore, enabling the circulation of the material resources contained in the SPA waste, would avoid the environmental problems associated with landfill sludge disposal, including metal lixiviation.

The NDSX/EW technology promotes the circular economy approach due to the recovery of several commercial products. Firstly, the generation of SPA in the HDG plants implies loss of metallic resources, specifically, between 5% and 6% of the primary zinc used in the molten zinc bath ends up in the SPA. Therefore, the recycling of zinc in the HDG process could improve the efficiency of zinc consumption in this sector. In addition, the recovery of iron chloride and a solution of sodium hypochlorite allows the obtention of products that could be reintroduced in the market. Secondly, auxiliary materials of the NDSW could be reused in the process. On one hand, the reutilization of the tap water used as stripping after the EW stage reduces the consumption of water in the NDSX and avoids the management of the stripping stream after zinc electrodeposition. Furthermore, the regeneration of the organic phase used as extractant allows its reuse in the NDSX reducing the waste streams generated by the NDSX/EW technology.

Finally, despite the benefits of the NDSX/EW technology, the main challenge of the technology is the difficulty in ensuring high quality of the recovered materials. Further research will be needed to determine the quality of the recovered zinc as a substitute for primary zinc in galvanizing processes. In addition to the environmental advantages, this alternative would be desirable because of its economic benefits, which will be influenced by the final quality of zinc, and the zinc price in the market. The other recovered products, iron chloride and sodium hypochlorite, would also be beneficial, but to a lesser extent compared to zinc. In any case, recovered zinc could still be used as a raw material for other sectors with lower quality requirements, such as in zinc oxide applications. Life cycle costing is recommended in order to search the trade-off between environmental and economic objectives.

Credit author statement

Andrea Arguillarena: Methodology; Investigation; Writing – original draft. **Maria Margallo**: Methodology; Formal analysis; Writing – review & editing. **Angel Irabien**: Formal analysis; Writing – review & editing. **Ane Urtiaga**: Conceptualization; Funding acquisition; Formal analysis; Writing – review & editing; Supervision

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2022.115567.

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