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Life Cycle Assessment of Cobalt Catalyst Production and Recycling

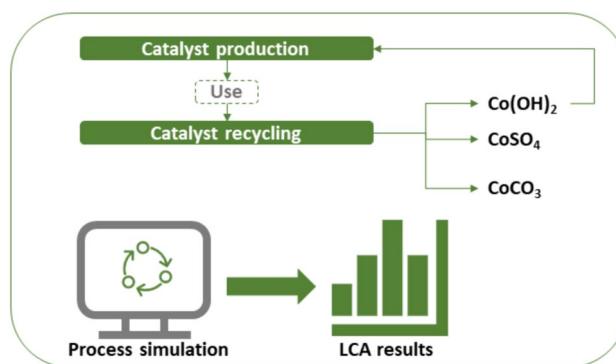
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Abstract

Catalysts with an active phase of cobalt are crucial for Fischer–Tropsch synthesis (FTS), yet the environmental impacts of the catalyst production and the recycling of the spent catalyst remain largely unknown. The goal of this study was to evaluate the impacts of both catalyst production as well as the recycling of spent catalyst as cobalt hydroxide, cobalt sulfate, or cobalt carbonate. Life cycle assessment (LCA) was used to quantify the environmental impacts of the studied processes. The life cycle inventory (LCI) was gathered based on the mass and energy balances of process simulations built on information available in the literature. The results show that compared to primary production of equivalent products, all studied recycling processes for spent catalyst decrease the environmental impacts by more than 50% in all investigated impact categories. For example, the global warming potential (GWP) of cobalt recovery from spent FTS catalyst as cobalt sulfate was 1.7 kg CO₂-eq./kg CoSO₄ whereas the corresponding GWP for primary production was 4 kg CO₂-eq./kg CoSO₄. The process hot-spots of recycling were found to be the production of the chemicals consumed, particularly sodium hydroxide and sulfuric acid, which together contributed between 64 and 95% of the total environmental impacts. LCAs on FTS have included the consumption of cobalt catalyst in the LCI using various approximations. The impacts calculated for the production of cobalt catalyst in this study were found to be markedly higher. The largest contributors included the production of materials for the precursor and support, as well as NO_x emissions and consumption of nitric acid.

Graphical Abstract



Keywords Life cycle assessment · Process simulation · Environmental impacts · Recycling · Fischer–Tropsch catalyst · Cobalt chemicals

Introduction

The recycling and recovery of cobalt are becoming increasingly vital due to cobalt's status as a critical raw material. The consumption of cobalt is expected to increase substantially as a result of its use in batteries, particularly those in

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Extended author information available on the last page of the article

electric vehicles, but its primary production is geographically concentrated in the Democratic Republic of the Congo and China with little extraction or refining elsewhere in the world [1]. The high economic importance of cobalt along with the possibility for supply disruption for political reasons, for example, indicates that diversification of its supply would be beneficial. In addition to primary production, cobalt can be recovered from many types of cobalt containing waste and scrap, including spent cobalt catalyst. Recycling of cobalt is important both due to the necessity of diversifying the supply of cobalt and the possibility for less environmental impact. The recycling of cobalt has been found to have smaller environmental impacts compared to primary production of equivalent products in the case of hardmetal [2] and batteries [3]. The impacts of cobalt recycling from spent FTS catalyst have not been evaluated.

Cobalt containing catalysts are used in two main applications: in the hydrodesulfurization (HDS) process to remove sulfur from oil, and in Fischer–Tropsch synthesis (FTS) to make synthetic fuel from natural gas [4]. Both HDS and FTS catalysts are heterogeneous catalysts that can be produced by impregnation of a metal precursor, which provides the catalytically active phase, onto a support that helps maximize the surface area of the catalyst. Activation is necessary to produce active material from the metal precursor. In addition to the active material and the support, catalysts may contain promoters depending on the desired catalyst characteristics. In HDS catalyst molybdenum is the active material and the main component by weight excluding the alumina support. They only contain 1–6% cobalt, which is added as a promoter [4]. In a cobalt-based FTS catalyst, the share of cobalt can be much higher than in an HDS catalyst [5]. Approximately 5% of the global consumption of cobalt is used for the production of cobalt catalysts, and this amount has been increasing in the past two decades. Cobalt salts can be used as precursor materials for industrial catalysts as cobalt is multivalent and enhances catalytic action. In an FTS catalyst, cobalt can be replaced by ruthenium or iron, but cobalt remains the preferred option as it provides the highest yield and longest lifetime [1].

The recycling rate of spent cobalt catalyst has been estimated to be quite low, approximately 5% [6]. The Cobalt Institute has estimated that the total recovery of cobalt from secondary raw materials was 10 600 t in 2020. Of this, 11% came from catalysts and alloy scraps. Considering that global production of refined cobalt in 2020 was 132 000 t, of which approximately 5% was used in catalysts, the recycling rate of cobalt catalysts appears to remain low [7]. Most efforts to recycle cobalt from spent catalyst are based on hydrometallurgical treatment, mostly by dissolution in inorganic acids [4]. Spent cobalt FTS catalyst can be recycled by using it as feed material in the same processes that are used to refine primary cobalt

raw materials [8]. Recycled cobalt as e.g., hydroxide or carbonate may then be used again for the production of catalyst.

Several life cycle assessment (LCA) studies have been performed for FTS processes. They include the impacts of cobalt catalyst used in the synthesis but only in limited detail due to a lack of comprehensive data. Iribarren et al. [9] studied production of Fischer–Tropsch products from biosyngas and included cobalt catalyst in the life cycle inventory (LCI). Navas-Anguita et al. [10] determined the environmental impacts of the production of synthetic fuels by biogas dry reforming and FTS. In both Iribarren et al. [9] and Navas-Anguita et al. [10], the background data were obtained from the ecoinvent database, and therefore, it is difficult to determine what is included in the catalyst production system as the database does not currently contain a process for cobalt catalyst production. Liu et al. [11] reported greenhouse gas emissions from direct air capture and Fischer–Tropsch fuel production and also included a cobalt catalyst in the LCI, but considered only metal production and not the steps for catalyst fabrication. Similarly, Rangel et al. [12] used the ecoinvent database for catalyst background data in an FTS process and included ore extraction and metals refining but not catalyst fabrication. Okeke et al. [13] studied the environmental impacts of renewable diesel production via anaerobic digestion and FTS. They used the GREET model [14] for catalyst LCI data. The GREET model reports LCI data for primary production of refined cobalt and selected cobalt chemicals. The GREET catalyst module contains data for the fabrication of five selected catalysts, one of them being Mo/Co/ γ -Al₂O₃, which is the only one containing cobalt.

In this study, the goal was to provide LCI data on cobalt recovery from spent FTS cobalt catalyst and subsequent catalyst production as well as to study the environmental impacts of the processes to identify hotspots in the systems. The recycling impacts were compared to the impacts of the primary production of equivalent products to determine the possible environmental benefit gained by recovering cobalt from the catalyst.

Materials and Methods

The simulation parameters, e.g., reaction yields and reactant concentrations, and the process chemistry for each unit process were determined based on laboratory and pilot-scale studies. Process simulations were conducted using HSC Sim 10 [15] based on the literature values and typical metallurgical production. Mass and energy balances from the simulations were used to compile an LCI and lastly, a life cycle impact assessment (LCIA) was conducted using GaBi software [16] and the ecoinvent 3.8 database [17].

Spent Catalyst Raw Material

The active phase in the supported cobalt catalyst used in FTS consists of metallic cobalt. The catalyst degrades eventually in use, affecting the active phase composition. When entering the recycling process, some of the cobalt probably exists as cobalt oxide or a mixed metal oxide, e.g., CoTiO_2 when the support is titanium dioxide. A mixed metal oxide might be formed as a result of an undesirably strong interaction between cobalt and the support leading to deactivation of the catalyst [18]. The most common supports in FTS catalyst are titanium dioxide, silicon dioxide, aluminum oxide, and carbonaceous material [5]. Promoters can be added to the catalyst to achieve the desired catalyst characteristics, e.g., to make the reduction temperature lower. The promoters used and studied include noble metals (e.g., Pt, Ru, Ir), transition metal oxides (e.g., Zr, Mn, Ni), and alkali and alkaline earth metals (e.g., Li, Na, Ca, Mg) [5]. Noble metals are used in low amounts, 0.1–0.5 wt%, due to both their high cost and the fact that higher amounts may lead to blockage of the cobalt active sites. The other promoters are also used mainly in fairly small amounts, ranging from 0.2 to 5 wt%, but may range up to 20 wt% for some transition metals [5].

The feed to the simulated recycling process consisted of a mix of FTS cobalt catalyst materials in equal amounts by mass. The promoter compositions varied and were based on possible FTS catalyst compositions detailed by Gholami et al. [5]; they are listed in Table 1.

Process Description

The catalysts used in FTS are heterogeneous catalysts and can be produced by impregnation of a metal precursor onto a support. Cobalt nitrate is a typical precursor, and it can be prepared from cobalt hydroxide or carbonate, for example, by treating them with nitric acid, after which cobalt nitrate can be crystallized [19]. Titanium dioxide is used as the support material in the catalyst production model. Impregnation of the precursor onto the support is followed by drying in a nitrogen atmosphere. Calcination and reduction are used to perform the activation to transform the metal precursor into the catalytically active form. The reduction in hydrogen is

necessary to achieve a catalyst where the cobalt exists in a metallic phase [19].

Spent cobalt FTS catalyst can be treated hydrometallurgically in the same refining process used for primary cobalt raw materials. The raw material is first leached in sulfuric acid to solubilize cobalt and other valuable metals. The solution is then purified in several steps including precipitation and solvent extraction. After purification the treatment of the solution depends on the desired end-product [8]. In the current study, three scenarios for cobalt recovery were assessed. Scenario 1 (SCE1) assessed cobalt recovery as cobalt hydroxide by precipitation from the purified solution using sodium hydroxide. Scenario 2 (SCE2) assessed recovery as cobalt sulfate by crystallization from the purified solution. Scenario 3 (SCE3) assessed recovery as cobalt carbonate by precipitation from the purified solution using sodium carbonate. The purity of the recovered cobalt products fulfills the chemical specifications for cobalt salts outlined by Sole et al. [20].

Waste management in all the assessed processes includes sulfate removal and neutralization of the produced wastewater before release. A schematic of the studied system is shown in Fig. 1. Detailed process descriptions and flowsheets are available in the online supplementary material in sections S1 and S2, respectively.

Goal and Scope Definition

The goal of the assessment was to conduct a gate-to-gate LCA of the environmental impacts of FTS cobalt catalyst recycling in a typical cobalt refining process and the cradle-to-gate impacts of FTS cobalt catalyst production. Simple cut-off was used as the end-of-life allocation method and recycling, so the cobalt chemicals produced from the spent catalyst was only considered responsible for the environmental impacts of the subsequent recycling process. LCI data for the foreground system of catalyst production and recycling was generated with the use of process simulation. Background processes including primary production of cobalt and the support in the catalyst were modeled using the ecoinvent 3.8 [17] database. The functional unit for the recycling process was defined as 1 metric ton (mt) of cobalt catalyst with a cobalt content of 20 wt-%. The functional

Table 1 Feed composition for the simulated process of cobalt catalyst recycling

	Composition 1	Composition 2	Composition 3	Composition 4	Composition 5
Co, %	20	20	20	20	20
TiO ₂ , %	80				
Al ₂ O ₃ , %		80		79	79
SiO ₂ , %			80		
Mn, %				1	
Ca, %					1

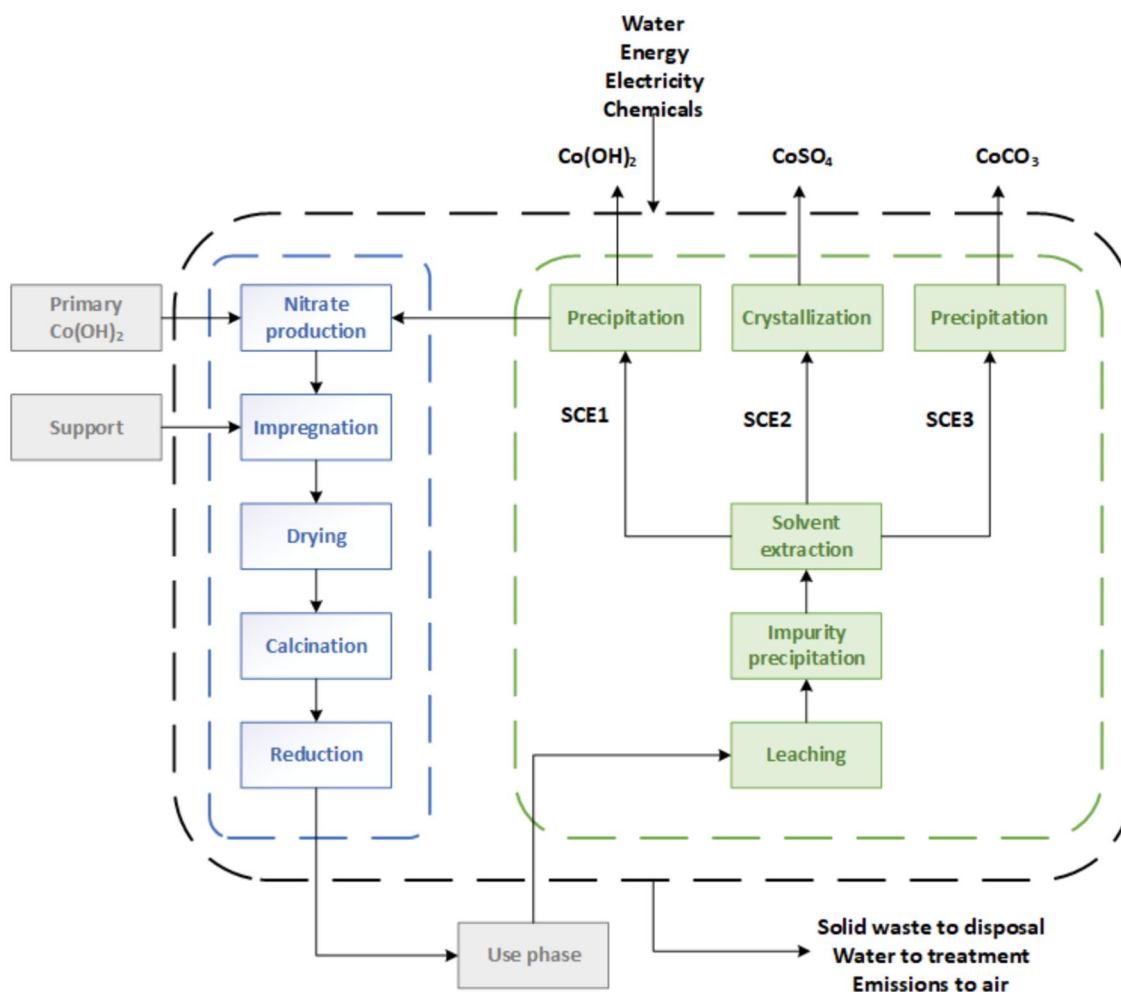


Fig. 1 Technical boundary used in the study for cobalt catalyst production and recycling represented by the black border. Catalyst production in blue, recycling of spent catalyst in green, and stages not

modeled in gray. SCE1 represents cobalt recovery as cobalt hydroxide, SCE2 recovery as cobalt sulfate, and SCE3 recovery as cobalt carbonate (Color figure online)

unit used for the catalyst production process was 1 mt of cobalt catalyst with a cobalt content of 20 wt% and titanium dioxide support.

The scope of the recycling process was limited to the recovery of cobalt. Other elements contained in the spent catalyst raw material were assumed not to be recovered in the process and were treated as waste in the LCIA. The amount of promoter metals contained is low and while it may be possible to recover titanium from the spent catalyst, not enough information on chemical behavior was available in the public literature to consider it as a scenario. The use phase of the catalyst was omitted from the assessment due to the lack of available data. Europe was considered as the geographical boundary and European values were used for the background processes whenever available. The system boundary is presented in Fig. 1.

The impact of using secondary and primary cobalt hydroxide for the production of the catalyst was studied by

constructing three scenarios. In scenario M (SCE (M)), the cobalt hydroxide feed used to produce the cobalt precursor for catalyst production was assumed to be a mix of 33% primary cobalt hydroxide and 67% secondary cobalt hydroxide from catalyst recycling. In scenario S (SCE (S)), the cobalt hydroxide feed was assumed to be 100% secondary cobalt hydroxide from catalyst recycling. In scenario P (SCE (P)), the cobalt hydroxide feed was assumed to be 100% primary cobalt hydroxide. The results of SCE1 were used to determine the environmental impacts of recycled cobalt hydroxide.

The impacts of the simulated recycling processes were compared to the impacts of producing equivalent amounts of each of the three cobalt products from primary raw materials. The comparison was done based on product mass. The impacts of primary production were gathered fromecoinvent 3.8 for cobalt hydroxide and cobalt carbonate, and from the Cobalt Institute [27] for cobalt sulfate. The environmental

impacts of cobalt sulfate production were only compared for GWP, EP, POCP, and AP, as the Cobalt Institute does not report values for ODP or FC.

LCIA was conducted using ReCiPe 2016 with a hierarchist perspective. The impact categories evaluated in this work were global warming potential (GWP, measured in kg CO₂-eq.), terrestrial acidification potential (AP, kg SO₂-eq.), freshwater eutrophication potential (EP, kg P-eq.), ozone depletion potential (ODP, kg CFC-11-eq.), photochemical ozone creation potential (POCP, kg NO_x-eq.), and freshwater consumption (FC, m³). The choice of evaluated impact categories was based on the recommendation of Santero and Hendry [21] and the fact that they well represent the environmental impacts on air, water, and soil.

The interpretation addressed three main topics: identification of significant issues, general comparison of the LCIA results of the different modeled scenarios, and provision of conclusions, limitations, and recommendations. The data used was evaluated for completeness, sensitivity, and consistency.

Inventory Analysis

Process simulation was used to generate the LCI data necessary to study the environmental impacts of the system. The methodology has been used previously for a variety of detailed studies on metallurgical processes [2, 3, 22–25]. Simulation parameters including process conditions, reagent concentrations, chemical consumptions, and extraction yields were gathered from the literature. These were mainly based on laboratory-scale processes, information provided in Finnish environmental impact assessment (EIA) procedures. When suitable simulation parameters were not available in the literature, they were estimated to the likely order of magnitude based on hydrometallurgical processes using similar unit operations for raw materials with similar valuable metal contents. The different levels of confidence in the input data are considered in the sensitivity and uncertainty analyses.

As most of the literature data used for the process simulation came from laboratory-scale studies, the parameters used were not fully optimized for an industrial-scale process. This may have resulted in higher chemical and water consumptions compared to an industrial process, although the recycling of water was included in the simulations. The process chemistry in the simulations was simplified but the actual chemistry may be more complex and for instance follow different dissolution reactions. The consumption of chemicals was also determined stoichiometrically, while in reality, the consumption and process conditions may be affected by the presence of impurities not reported in the public literature. The uncertainty in the LCIA results due to the use of process simulation has been considered in the interpretation and formulation of conclusions and recommendations.

The mass and energy balances of the process simulations were used to list the inputs and outputs for the processes, and these were used to form the LCI. The outputs from the simulations included consumption of chemicals and raw materials, emissions to air and water, and the amount of generated waste. The electricity consumption of reactors, thickeners, filters, and crystallizers was also calculated based on the process simulations. The calculation method for the electricity consumption of the unit processes has been previously detailed by Elomaa et al. [22] and Rinne et al. [3]. As the processes also consume energy and electricity in other parts of the process, additional information was also utilized. For the recycling processes, the information was collected from two cobalt producer EIAs [8, 26]. The additional energy and electricity consumption of catalyst production was estimated based on the GREET catalyst module [14]. Nitrogen dioxide is produced during the calcination step of catalyst production and flue-gas cleaning was assumed using addition of ammonia, which reacts with nitrous oxides to produce nitrogen and water vapor. Ammonia consumption was estimated based on stoichiometry due to the lack of primary data. The flowsheets used in this work are shown in the online supplementary material in Figs. S1–S4 for SCE1, SCE2, SCE3, and catalyst production, respectively. The parameters and their sources have been detailed in the online supplementary material Tables S1–S4 for SCE1, SCE2, SCE3, and catalyst production, respectively. The final LCI data for the studied system is presented in Table 2.

Results

Catalyst Production

The LCIA and contribution analysis results for catalyst production are shown in Fig. 2. The largest contributors to the environmental impacts of catalyst production are mainly the production of cobalt hydroxide and titanium dioxide, the resources required for the catalyst itself. Even when only secondary cobalt hydroxide is used, the contribution is notable, i.e., between 18 and 42% of the total impacts. When only primary cobalt is used, cobalt hydroxide is the single largest contributor in all investigated indicators contributing 52–95% of the total impacts. Of the remaining impacts, a substantial share is due to the production of titanium dioxide, the contribution of which is 26–89% depending on the impact category.

The impacts from the production of the chemicals and energy used in the process are mostly minor. POCP and ODP form the exception. For POCP, a large contribution comes from direct emissions, which account for 28–47% of the impacts depending on the source of the cobalt hydroxide that is used in the catalyst production. The impact of

Table 2 Life cycle inventory for production of 1 mt cobalt catalyst and recycling of 1 mt spent cobalt catalyst into cobalt hydroxide (SCE1), cobalt sulfate (SCE2), and cobalt carbonate (SCE3)

Catalyst production			
Inputs			
Cobalt hydroxide, mt	0.36		
Water, mt	5.8		
Support, mt	0.79		
Electricity, kWh	2400		
Heat, kWh	190		
Hydrogen, mt	0.010		
Nitric acid, mt	0.45		
Carbon dioxide, mt	1.3		
Ammonia, mt	0.11		
Outputs			
Offgas, mt	5.4		
Activated catalyst, mt	1.0		
Treated effluent, mt	2.7		
Solid residue, mt	0.050		
Recycling	SCE1	SCE2	SCE3
Inputs			
Scrap, mt	1.0	1.0	1.0
Water, mt	7.9	14	8.0
Sulfuric acid, mt	1.7	2.2	1.7
Hydrogen peroxide, mt	0.0096	0.0096	0.0096
Electricity, kWh	900	950	900
Heat, kWh	800	800	780
Air, mt	0.019	0.019	0.019
Sulfur dioxide, mt	0.0016	0.0016	0.0016
Calcium oxide, mt	0.15	0.55	0.14
Sodium hydroxide, mt	0.81	0.49	0.66
Carbon dioxide, mt	0.032	0.028	0.025
Kerosene, mt	0.015	0.015	0.015
Extractant, mt	0.0038	0.0038	0.0038
Flocculant, mt	0.031	0.031	0.031
Sodium carbonate, mt			0.23
Outputs			
Solid residue, mt	4.9	5.8	4.9
Offgas, mt	0.032	6.6	0.032
Cobalt product, mt	0.25	0.71	0.32
Treated effluent, mt	6.6	4.8	6.7

the direct emissions is mostly due to the nitrogen dioxide formed during calcination even with abatement assumed. For ODP, a large contributor is the production of the nitric acid used for the production of the cobalt nitrate precursor from cobalt hydroxide. Nitric acid accounts for 20–46% of the total ODP impacts.

The results show that the environmental impacts of secondary cobalt hydroxide production are substantially smaller than those for the production of an equal amount from primary sources. The impacts of secondary production

are between 71 and 96% smaller than those of primary production depending on the impact category. The difference is largest for FC and smallest for AP. As the production of cobalt hydroxide is a major contributor to all investigated impact categories, significant reduction in the impacts of total catalyst production is possible when the share of secondary cobalt is increased. SCE (M) with a mix of primary and secondary cobalt results in a decrease in catalyst production impacts of 26–61% compared to SCE (P) when only primary cobalt is used. SCE (S) using only secondary cobalt

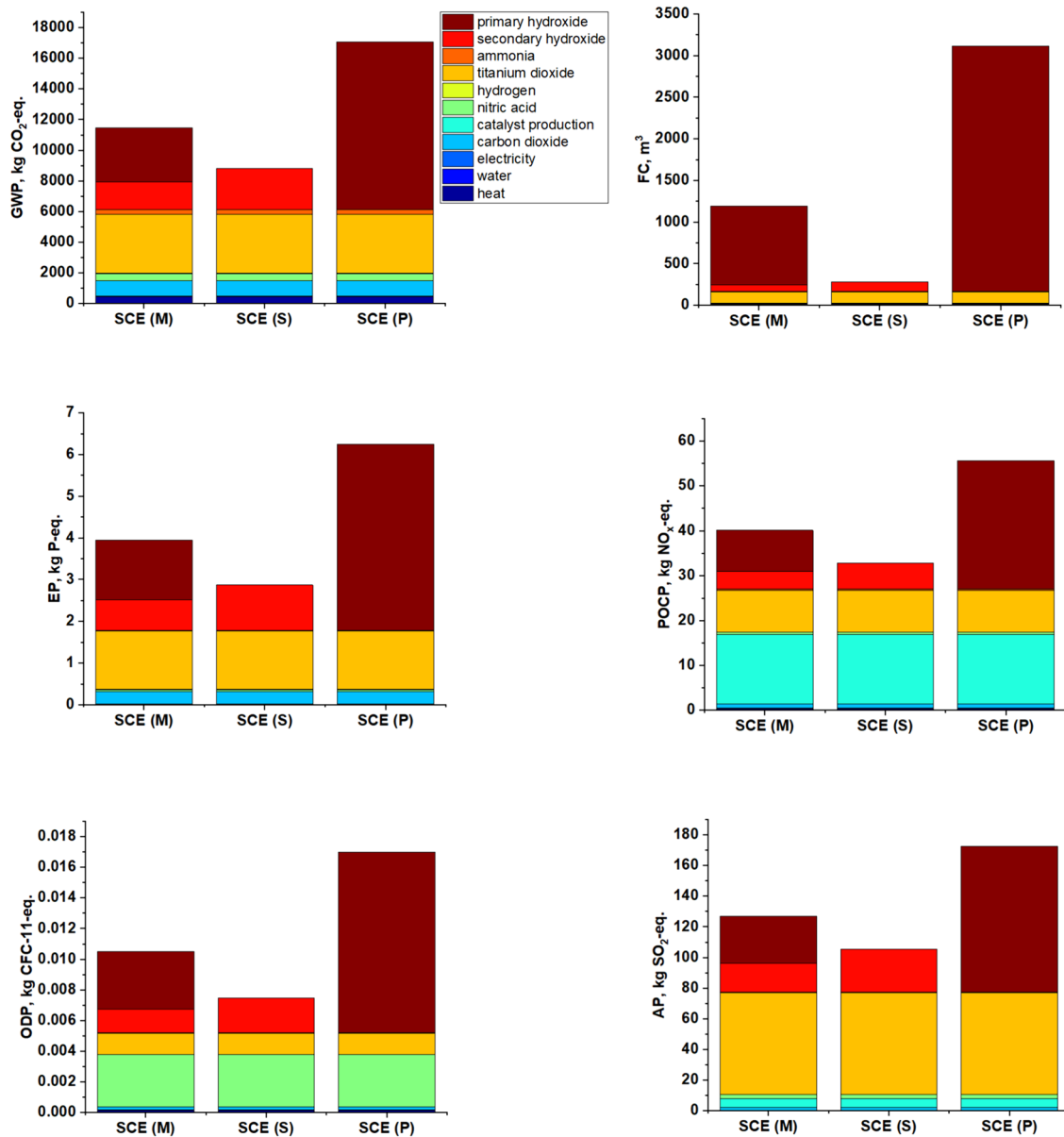


Fig. 2 Environmental impacts of production of 1 mt cobalt catalyst when cobalt hydroxide comes from recycling of spent catalyst supplemented by primary production (SCE (M)), all from secondary production (SCE (S)), and all from primary production (SCE (P))

decreases the impacts even further, by 39–91% compared to SCE (P).

Spent Catalyst Recycling

The LCIA results for recycling of spent catalyst into different cobalt products are shown in Fig. 3. The results show that the impacts of the recycling processes are mainly due to the production of the consumed chemicals and utilities and the direct impacts of the processes themselves are negligible.

The largest contributors for each of the indicator categories are similar for each of the recycling scenarios. The

production of sodium hydroxide is the largest contributor for GWP, EP, POCP, and ODP, whereas the production of sulfuric acid is the largest contributor for FC and AP. The single largest contributor contributes between 44% (SCE1, FC) and 83% (SCE2, AP) of the total impact depending on the scenario and indicator. Together, the production of sodium hydroxide and sulfuric acid contributed between 64 and 95% of the total environmental impacts. The impacts of the different scenarios are in a similar range for all indicators. For FC, differences between the total impacts of the scenarios are small. For GWP, EP, POCP, and ODP, the impacts appear to be the highest for cobalt

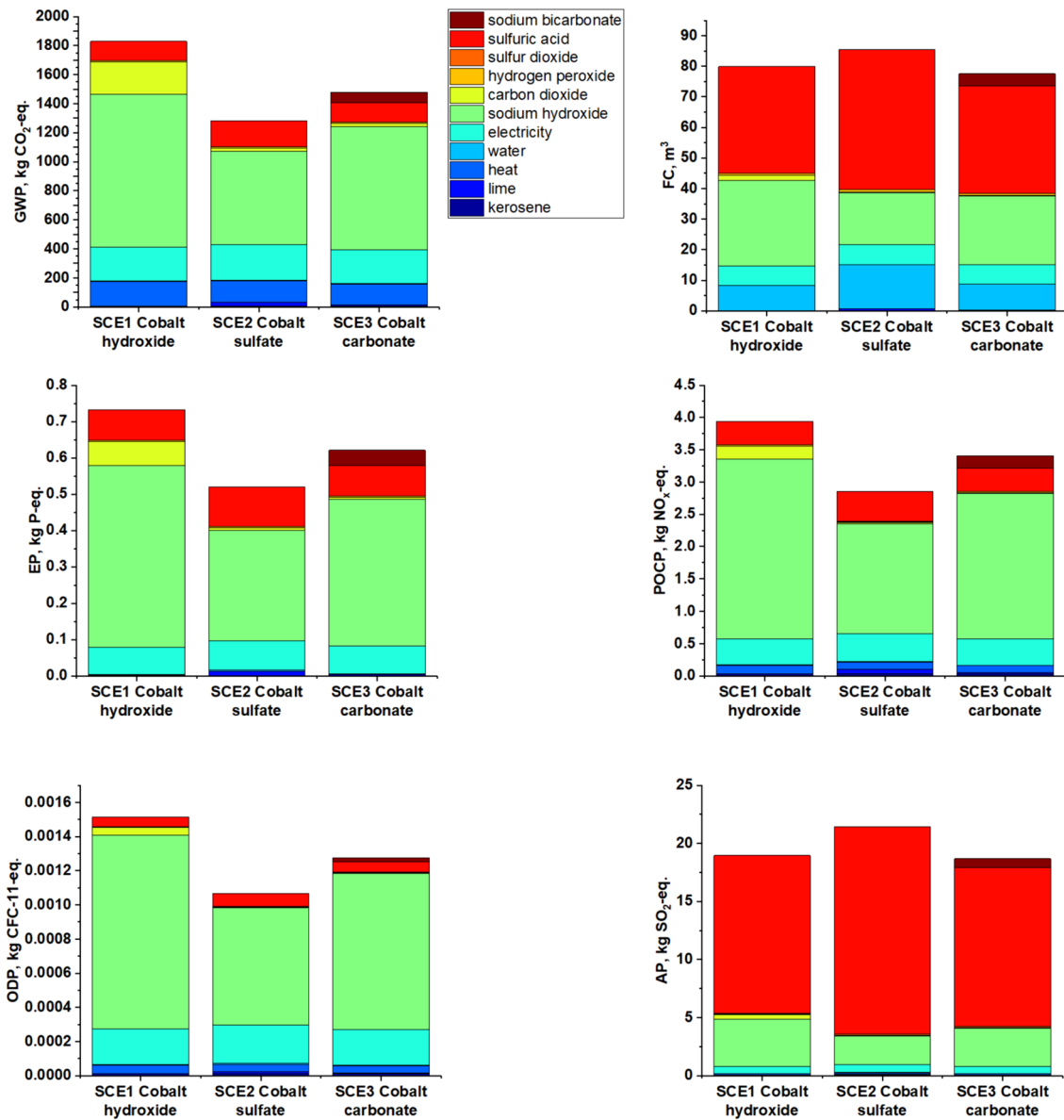


Fig. 3 Evaluated environmental impacts of production of recycling of 1 mt spent catalyst into cobalt hydroxide, cobalt sulfate, and cobalt carbonate

hydroxide production and smallest for cobalt sulfate with the difference being 27–30%.

The impacts of the simulated recycling processes were compared to the impacts of producing equivalent amounts of each of the three cobalt products from primary raw materials. The results of the comparison are shown in Fig. 4, where the impacts have been scaled so that 100% represents the value of primary production impacts for each cobalt product and impact category. The results show that the environmental impacts of producing recycled cobalt products from spent catalyst are less than 45% of the environmental impacts of equivalent products from primary raw materials, for all indicators and products. The recycling impacts are mostly less

than 25% of the primary production impacts, apart from GWP for cobalt sulfate and AP for cobalt hydroxide and cobalt carbonate.

Sensitivity and Uncertainty

A sensitivity analysis was conducted to study the sensitivity of the LCIA results to variation in the values of the individual items that form the LCI. The sensitivity was studied by varying the values of the LCI items in Table 2 individually by $\pm 20\%$ in GaBi and then recalculating the LCIA results. The new LCIA results were then compared to the original LCIA results to identify which impact categories were

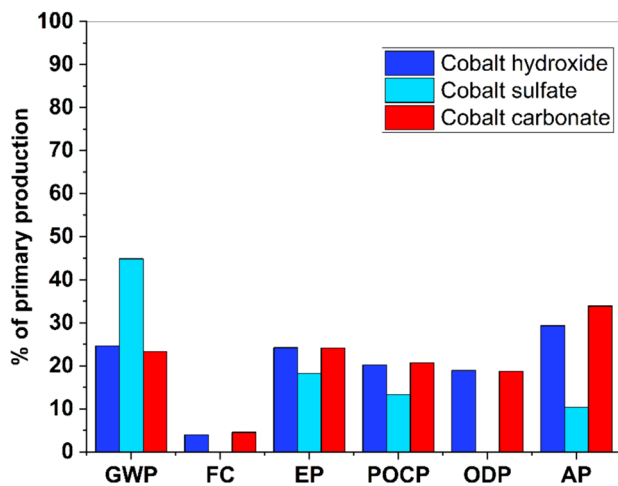


Fig. 4 Evaluated environmental impacts of cobalt recovery as hydroxide, sulfate, and carbonate from spent cobalt catalyst compared to primary production. Impacts have been scaled so that the impacts of the primary production of each product equal 100%

substantially affected by the change. The amount of 20% was chosen as the change in the original values as it was considered to be substantial enough to show possible sensitivity in the LCA model and appropriate for the level of uncertainty in the simulation data which is based mainly on the literature. The LCI items studied in the analysis included all the chemicals and utilities consumed. Of the direct emissions, only the nitrogen dioxide from the catalyst production was included as the impacts from other direct emissions were negligible. Fixed values determined by the functional unit such as the amounts of catalyst precursor and support were not included in the varied LCI items.

The recycling models in SCE1, SCE2, and SCE3 were mainly sensitive to the value of sodium hydroxide and sulfuric acid consumption. A decrease in sodium hydroxide consumption decreased the environmental impacts by 2–15% depending on the scenario and indicator. Similarly, an increase in consumption increased the environmental impacts by 2–15%. The effect was most pronounced (> 10%) in GWP, EP, POCP, and ODP. The sulfuric acid consumption mainly affected FC and AP, although the impact was more pronounced in AP. A reduction in consumption decreased the impacts by 9–17% and similarly an increase in consumption increased the impacts by 9–17%.

The catalyst production model was mainly sensitive to the consumption of nitric acid and NO_x emissions from calcination. Nitric acid consumption affected the ODP, i.e., a decrease in consumption decreased the impacts by 13% and an increase in consumption increased the impacts by 13%. NO_x emissions affected the POCP so that a decrease in emissions decreased the impacts by 12% and an increase in emissions increased the impacts by 12%.

An uncertainty analysis was also conducted to provide information on the reliability of the conclusions and the recommendations drawn from the results. Data quality indicators with corresponding levels of uncertainty were assigned to the LCI items from the sensitivity analysis; they are presented in detail in the online supplementary material Tables S5 and S6. A Monte Carlo analysis was performed in GaBi software with 100 runs and normal distributions. For catalyst production, the standard deviations were mainly small, between 1 and 3%. POCP and ODP formed the exceptions, for which the standard deviations were 8% and 13%, respectively. For the recycling models the standard deviations varied between 6 and 11%, mostly being highest for AP.

Discussion

The environmental impacts of catalyst production were studied in this work. A substantial amount of the environmental impacts was found to be contributed by the production of the materials required for the catalyst, namely titanium dioxide used as the support and cobalt hydroxide used for the production of the cobalt precursor, especially when using only primary cobalt hydroxide. Also, direct NO_x emissions and consumption of nitric acid made a substantial contribution to POCP and ODP, respectively. The GWP per kg of produced catalyst was found to be 8.8 kg CO₂-eq. when using only secondary cobalt, 11.5 kg CO₂-eq. when using a mix of secondary (67%) and primary (33%) cobalt, and 17.1 kg CO₂-eq. when using only primary cobalt. Iribarren et al. [9] found that in the production of Fischer–Tropsch products from biosyngas, the production of the cobalt catalyst used in FTS dominates most impact categories gate-to-gate, e.g., accounting for approximately 55% of the GWP. The GWP determined by Iribarren et al. [9] was 3.70 kg CO₂-eq. and the system included 0.27 kg of catalyst, which translates to a GWP of roughly 8 kg CO₂-eq./kg catalyst. It seems likely that approximating the contribution of cobalt catalyst and not including the contribution of the catalyst production process and all its elements may substantially underestimate the environmental impacts. Generic impact data of a cobalt production process cannot accurately describe the impacts of cobalt catalyst production or recycling and more detail should be included in the system when possible.

Recycling of spent catalyst and its environmental impacts were also studied, and the modeled recycling process was based on the unit processes used in industrial cobalt refining. It was found that the production of the chemicals consumed was the main contributor, with the production of sodium hydroxide and sulfuric acid in particular accounting for the majority of the environmental impacts. The gate-to-gate GWP for the production of 1 kg cobalt sulfate from spent cobalt catalyst in the current study was 1.7 kg CO₂-eq. The

Cobalt Institute has also provided LCIA results for several cobalt products, including cobalt sulfate for which they report a GWP of 4.0 kg CO₂-eq./kg product cradle to gate, with the refining stage accounting for 2.0 kg CO₂-eq. [27]. In spite of not directly comparable due to different system boundaries and production processes, the magnitudes of GWP for cobalt sulfate production were found to be in the same range. Similar to the current study, the Cobalt Institute also found auxiliaries such as chemicals consumption to be the largest contributor in the refining of cobalt as cobalt sulfate [28].

As the production of the consumed chemicals was the main contributor in the recycling process, the recycling impacts could possibly be further decreased by sourcing the main contributing chemicals from areas where the environmental impacts of production are lower. The production of sodium hydroxide is substantial contributor in the refining of cobalt. It is produced by chlor-alkali electrolysis in which a marked share of the environmental impacts is due to the production of the electricity consumed in the process [17]. Use of sodium hydroxide produced in areas with high green energy capacity could also decrease the environmental impacts resulting from cobalt refining.

Based on the indicators studied, recycling of cobalt from spent catalyst was found to be environmentally beneficial when compared to the production of equivalent products from primary raw materials. The recycling impacts were mostly less than 25% when compared to the primary production impacts. The only exceptions were GWP for cobalt sulfate production and AP for cobalt hydroxide and cobalt carbonate production, although these were still less than 45% of the primary production impacts. A substantial decrease in the environmental impacts can be achieved when cobalt hydroxide, cobalt sulfate, or cobalt carbonate produced from primary sources is replaced by production from secondary resources, including spent catalyst.

With the cobalt industry and its customers already aware and becoming increasingly conscious of the environmental footprints of products, the use of recycled cobalt is likely only to increase. However, the possible benefit offered by the recovery of cobalt from spent catalyst can only be realized by efficient collection schemes to increase the amount of spent catalyst being recycled. Closed loop recycling is an example where the cobalt-bearing materials used by customers are returned to the cobalt refiner for recycling. Projects to commercialize recycling of new cobalt-bearing materials is the subject of ongoing research interest [29]. Cobalt recycling can benefit from a tailored approach to enable full potential recovery in an environmental manner as the secondary raw material compositions and structures are varied.

The tools, data sources, assumptions, and modeling choices used in the study contribute to uncertainty and

affect how the results should be interpreted. The process models were simulated as industrial scale, but the process parameters were mostly based on laboratory-scale data. Using process simulation as a tool to generate LCI data can lead to small underestimation of environmental impacts, but it is more likely to overestimate impacts in some impact categories [28]. Compared to a laboratory-scale batch process, an optimized industrial process may perform better and for example, achieve higher yields, and recycle water and chemicals more efficiently. Some of these qualities such as recycle streams are also included in the upscaled process simulation while others, such as recoveries, are more difficult to estimate.

The uncertainty from different sources has been considered in the interpretation of the results of the LCA. This study provided gate-to-gate LCI data on spent cobalt catalyst recycling and catalyst production in the EU. The data can be used to estimate with reasonable accuracy the environmental impacts of processing cobalt raw materials of similar composition and production of cobalt catalyst with titanium dioxide support in the absence of primary plant data. However, adjustment of the data may be necessary if the composition of either the treated cobalt material or produced catalyst is significantly different to those used in the current study. Considering the uncertainty, the results of the different recycling scenarios cannot be easily distinguished in a meaningful way as this would also depend on the final application. In comparison to primary production, however, the environmental impacts of all recycling scenarios were substantially smaller than production of equivalent products from primary raw materials. Therefore, the results are encouraging for the recycling and recovery of cobalt from spent FTS catalyst. With suitable data available, the recovery of other valuable metals in the raw material, such as the support, should also be examined as to whether it would enhance the recycling system performance further in comparison to primary production.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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