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Material sustainability evaluation of electrocatalysts in early-stage research – case study on $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ in alkaline water electrolysis

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ABSTRACT

Sustainable energy materials for energy storage applications are crucial for a reliable energy transition. However, evaluating material sustainability in early stages of energy materials research is a big challenge, as it is difficult to predict the final industrial process and supply chain of the investigated materials. In this work we showcase that a sustainability evaluation can already be conducted in the early stage of electrocatalyst research. We provide a case study of two highly active alkaline oxygen evolution catalysts, $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$, with a thin film model-catalyst approach. For the sustainability evaluation, we consider the material criticality, compare precursor prices, conduct an environmental impact analysis of the precursors based on life cycle assessment and provide a qualitative discourse for social impacts of raw material extraction. We find that $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ has a reduced environmental footprint, smaller precursor supply risk and 10 times lower precursor price compared to $La_{0.6}Sr_{0.4}CoO_{3-\delta}$. However, for both perovskites the high environmental impact and high supply risk of lanthanum is of immediate concern towards larger scale application purposes. Sustainability evaluation in the early stages of energy materials research can be a great opportunity and guidance to identify crucial bottlenecks and to develop new strategies for material selection. This can have a broad impact on later stages of research and development for the energy transition.

1. Introduction

Countries around the world attempt to reduce their dependence on coal, oil and gas in the energy, industry, and transportation sector [1]. Renewable energy generation, energy storage and conversion technologies are mandatory for a reliable energy transition and a proceeding defossilization of the economy. With that, the global economy develops from a fossil fuel-intensive to a material-intensive economy, as large quantities of complex functional materials are required for technologies such as solar panels, wind turbines, electrolyzers, fuel cells and batteries [2–4]. It is estimated that for fulfilling the Paris climate agreement, the mineral requirements will quadruple by 2040 compared to today's mineral needs in the energy sector [2,3]. The shift to material-intensive large-scale applications leads to new criticalities of raw materials, and technological, scientific, economic, environmental, and social challenges.

Energy storage and conversion technologies such as batteries, fuel cells and electrolyzers are particularly crucial for capping energy supply and demand gaps in the energy grid and are required for several power-to-X applications [5–7]. For example, water electrolysis such as alkaline water electrolysis (AWE) is a key pillar in the transformation process towards a defossilized economy. The hydrogen produced from renewable sources can be used directly as final energy carrier or can be converted further together with captured CO_2 into e.g. methane, synthesis

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gas, liquid fuels, or other chemicals [5,6]. The battery, fuel cell and electrolyzer technological core components typically consist of at least a cathode, an anode, and a solid or liquid electrolyte. Typical elements required for these core components include noble metals such as iridium (Ir), ruthenium (Ru) and platinum (Pt) for electrolyzers and fuel cells [8,9]. Furthermore, elements such as manganese (Mn), iron (Fe), cobalt (Co) and nickel (Ni) are used for the cathode in lithium-ion batteries [10] and solid oxide fuel cells [11], and in alkaline water electrolyzers for both electrodes [8]. Most of these elements are considered critical for the global economy. For example, the EU categorizes several of the materials shown in Fig. 1a critical [12] (see red colored materials), highlighting that key materials for electrolyzers, fuel cells and batteries are linked to a possible vulnerability of large economies such as the EU [12].

To limit or substitute those elements of concern, the early-stage materials science community makes large efforts to shed light on the chemical and physical properties of energy materials that are “less scarce”, “more earth abundant” and “less costly” than state-of-the-art electrolyzer, fuel cell and battery materials [13–19]. However, different terms such as “shortage”, “scarcity”, or “earth abundance” and the price are often not clearly defined and not quantified for the investigated materials. Moreover, the environmental, geopolitical, or social impacts are seldomly considered, although they are of high importance for the prevention of potential supply chain disruptions, ecosystem damage or social disparities. In general, early-stage research is not and should not be limited by selective material choices, as only unrestricted research across the full periodic table can uncover groundbreaking findings of natural phenomena and material properties. At the same time, a more holistic material evaluation in early-stage materials research presents an opportunity for sustainable energy materials research and development from the onset.

For this reason, there are growing efforts by governmental and scientific institutions to quantify potential risks for material supply, ecological, economic and social aspects as well as technological requirements for new electrolyzers, fuel cells and batteries [12,20–29]. Recently, the effort for holistic material choice evaluation in early-stage material development was recommended by Klemenz et al. who explain that a “second-best” performance material can be more sustainable in a holistic evaluation than the material with the higher performance [23]. Helbig et al. recommend to evaluate material sustainability in basic research to estimate possible supply risks and environmental impacts using the example of yttrium manganese oxide ($YMnO_3$) for electronic applications [24]. Porzio and Scown recommend to compare new battery technology performances and their potential environmental impact to push sustainable battery development [25]. Eikeng et al. provide an extensive study on the future availability of critical raw materials for commercial hydrogen electrolyzer and fuel cell technologies. They point out the potential supply lacks of, for example, Ni for AWE or platinum group metals for proton exchange membrane electrolyzers [30]. Yet, this study includes neither an estimate of new or alternative materials that are in early-stage development nor a full sustainability evaluation. However, conducting a more holistic material evaluation in early-stage materials research is extremely challenging because the observed material's functionality as well as physical and chemical properties in laboratory scale model systems need to be projected to global material supply chains and industrial application standards.

Here, we showcase how a more holistic material sustainability evaluation can be conducted in early stages of electrocatalyst research. First, we briefly survey important sustainability parameters and their evaluations applied for energy materials, because the know-how and implementation of different sustainability parameters is not yet common practice for energy materials research in early stages [23,24,26]. Based on this, we provide a case study where we compare the two perovskite oxides $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ as electrocatalysts for the oxygen evolution reaction (OER) in AWE with a model catalyst approach.

We choose perovskite oxide ABO_3 catalysts as they are often discussed as attractive candidates for low temperature AWE catalysts at the anode [18,31–38]. Perovskite oxides crystalize in an ABO_3 structure where the A-site is typically occupied by rare earth elements or alkaline earth elements and the B-site by transition metals (Fig. 1b) [39]. Their variety of physical and chemical properties [39] makes them also suitable for many applications such as solid oxide fuel cells [11,40,41] or high temperature water electrolysis [42,43]. Fig. 1b shows a sketch of an integrated AWE stack with the perovskite oxide OER catalyst layer that is marked in red. In AWE, perovskite oxides may improve the sluggish

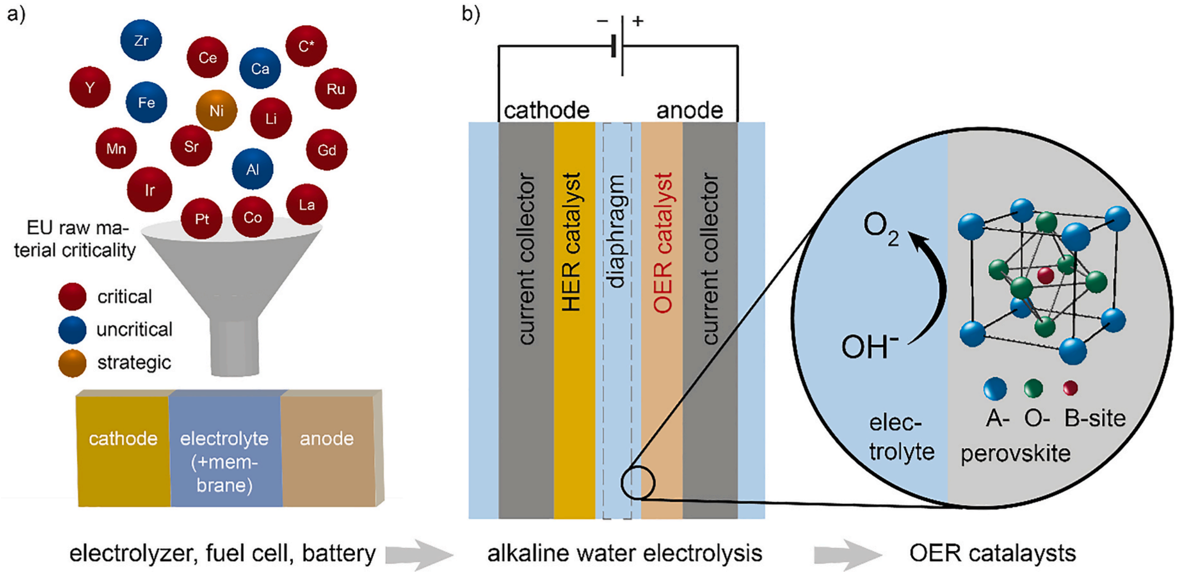


Fig. 1. a) Elements that are typically used for electrolyzers, fuel cells and lithium-ion batteries for the anode, cathode, electrolyte and membrane. The elements in red are considered critical in the EU criticality assessment 2023, the elements in blue are considered non-critical. Ni is a strategic element and Carbon (C*) is natural graphite [12]. b) Integrated AWE stack: anode and cathode consist of a current collector and catalyst layer. On the cathode, the hydrogen evolution reaction (HER) takes place and on the anode, the OER is catalyzed by the perovskites. The zoom on the right side shows the OER at the perovskite oxide/electrolyte interface and the perovskite crystal structure.

kinetics of the OER [31,32], in which hydroxyl ions are oxidized to oxygen gas at the perovskite catalyst interface (Fig. 1b).

$La_{0.6}Sr_{0.4}CoO_{3-\delta}$ is a widely studied OER catalyst exhibiting high electrocatalytic activity in AWE [33,35,44–46], (similar to the record-activity but structurally unstable ($Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$) [37,38,47,48]). However, strontium (Sr) and Co are elements of high concern due to their expected depletion of known reserves in the next 20-50 years and 50-100 years, respectively [49]. In contrast, calcium (Ca) and Fe are considered to have unlimited supply in a foreseeable future [23,49,50] and some studies have shown that $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ shows catalytic activity in AWE [37,51]. To compare the catalytic activity of the two perovskite oxides in the OER we take a thin film model catalyst approach as it facilitates the determination of intrinsic catalytic activity and leads to a reliable comparability of the catalyst performance as a basis for an early-stage sustainability evaluation [52].

The case study is applied to assess both electrochemical activity as well as material sustainability with material criticality, economic, environmental and social parameters. For the sustainability evaluation, we consider the electrocatalyst material criticality based on the critical raw material study of the European Commission [12]. Furthermore, we compare precursor prices of the two catalysts and consider several environmental impacts based on life cycle assessment (LCA). Additionally, we provide a qualitative discourse for social impacts of raw material extraction. The case study reveals that the use of Ca and Fe can indeed yield significant improvements in terms of material criticality, precursor prices and environmental impact. At the same time, however, our assessment also reveals that the material criticality and negative environmental impacts of lanthanum (La) is of concern for both perovskites. Our study highlights the necessity and chances to define and quantify sustainability parameters in early stages of energy materials research. At the end of the case study, we will then point out which information and initiatives can facilitate a more holistic material choice in the materials science community in the future.

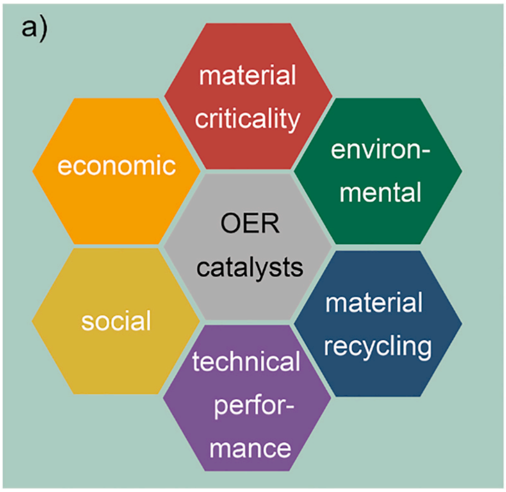
2. Methodology for a sustainability evaluation of energy materials

Fig. 2 displays the aspects of a sustainability evaluation of materials which are relevant for electrolyzers, fuel cells and batteries [23]. These sustainability evaluation concepts comprise material criticality, economic, environmental and social impacts, as well as material recycling and technical performance. As in the early stages of materials research the know-how and implementation of such sustainability parameters is not common practice [23,24,26], we briefly introduce the aspects shown in Fig. 2 and introduce which methods can be used in sustainability evaluations to analyze and quantify the different parameters. Each aspect contains a section on the methodology that was used in our case study for the evaluation of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ electrocatalysts, as summarized in Fig. 2b. For the case study, we first obtain the OER catalytic activity of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ by cyclic voltammetry (CV) to define the underlying base for the sustainability evaluation. Then we elaborate potential industrial synthesis routes to find common precursor materials that can be used for the fabrication of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ on large scale. Based on the catalytic activity and identification of the catalyst precursor materials, we determine the different sustainability parameters.

2.1. Material criticality

It is estimated that sufficiently large material resources exist for most of the currently used energy materials that are required for the full energy transition [2,3,53–55]. As resources are in definition economically viable extractable minerals today or in the closer future [56], it implies that the minerals earth abundance is high enough for the energy transition. However, despite the existence of sufficiently large resources, the current extraction rates and market supplies of some crucial materials are predicted to be too low to meet the extraordinarily increasing demand in material supply for the energy transition and other industries in the near future [2,3,53–55]. Hence, the material supply for the energy transition depends on the expansion of the mining industry and reliable supply chains within a network of geopolitical interdependencies, rather than on the general abundance of elements in the Earth's crust as a purely geological concept. Reviewing the current extraction and production rates [57] also in dependence of the depletion of known reserves [49] might give an overview of possible short-term material shortages, but neglects for example low supply chain diversifications, political instabilities of supplying countries, import reliance on other countries, export prohibitions, material recycling rates, or possible material substitution options in applications [12,27,49].

To evaluate these aspects, so-called material criticality assessments are used. The strongest economies USA [27,58], China [59], the EU [12] and Japan [60] periodically conduct criticality assessments for raw materials [61] to estimate material supply risks in their economy and to assess which impact a potential material shortage can have on their industrial sectors. For example, the critical raw material assessment published by the European Commission is based on the quantification of the EU material supply risk and economic importance in the most relevant industrial sectors [28]. Above a certain threshold of both



b)

Sustainability parameters	Methodology and resources
Material criticality	European Commission critical raw materials (EC-CRM 2023)
Economic	Precursor price comparison
Environmental	Precursor environmental impact based on LCA
Social	Qualitative indicators from literature
Technical performance	Cyclic voltammetry (CV) with perovskite oxide model catalysts
Material recycling	End-of-life recycling input rate (EOL RIR)

Fig. 2. a) Sustainability parameters relevant for a holistic evaluation of energy materials in electrolyzers, fuel cells and batteries with the example of OER catalysts in AWE. b) methods and resources used in the case study for the sustainability evaluation.

parameters, the material is defined as critical [12]. Because material criticality is a quantified and periodically updated parameter from the governmental institutions, the material criticality can serve as an instructive input to compare possible material shortages for energy materials in the early stages of energy materials research [26].

In this work, the European Commission's critical raw material methodology (EC-CRM 2023 [12]) was chosen to compare the material criticality based on the economic importance and supply risk of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ raw materials. It should be noted that the EC-CRM 2023 method is not a forecasting tool but uses recent data in its current CRM report [12]. The economic importance of a raw material is determined by the share of an end-use application's value added to the EU economy. It is reduced when the material in question can be substituted by another raw material in the end-application. The supply risk is calculated from the supplying country concentration weighted with the world governance indicators and also weighted with the import reliance of the EU. The supply risk is reduced by the end-of-life recycling input rate (EOL-RIR) and is also reduced when the raw material can be substituted by other materials in an end-application. The underlying calculations for the economic importance and the supply risk are shown and explained in detail in the supporting information (SI) supplementary note N1 and were taken from Ref. [28]. Moreover, we projected the possible demand of perovskite oxide precursors in AWE (based on our experimental results) in 2050 to estimate if supply bottle necks might occur for the EU economy.

2.2. *Economic parameters*

To predict the possible profitability of, for example, batteries, fuel cells or electrolyzers including their required materials, the investment and operational costs need to be related to the final market price. To calculate the full costs typically the CapEx (Capital Expenditure) and OpEx (Operational Expenditure) method are used [23,62,63]. To render the full life cycle costs of a technology the life cycle costing (LCC) method is a strong tool that can also include externalized costs of environmental impacts [23,64].

During early-stage materials research, the projected industrial CapEx and OpEx as well as market price are hardly known. However, there are possible options to estimate different costs. For example, the price of necessary precursor materials or the energy costs for material fabrication can be estimated if the (scalable) production process is projected.

In this work, we employ the precursor prices determined for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ to identify possible differences in economic impact. For precursor materials that are traded globally and have global market prices, the prices were taken from the institute of rare earth elements and strategic metals (ISE - Institut für Seltene Erden und Metalle) (see price details in SI Table T1 including the access date). For domestically traded precursor materials, price estimates were taken or requested from German suppliers (SI Table T1). A cumulative price of the precursor materials for 1 kg of stoichiometric $La_{0.6}Sr_{0.4}CoO_3$ and $La_{0.6}Ca_{0.4}FeO_3$ was calculated based on the required masses of the precursors shown in Table 1.

2.3. *Environmental parameters*

Despite being envisioned as environment-friendly technologies, the production, operation and end-of-life of electrolyzers, fuel cells and batteries can have severe impact on the environment, particularly given the high material demand [53]. Therefore, the minimization of environmental impacts must be considered over the entire life cycle of the technologies [2]. Especially, the mining and processing industries are extremely energy and resource-intensive and largely influence the environment [65]. For example, up to 2000 m^3 of water are required to extract one ton of Co metal, which even increases up to 1.2 Mio m^3 of water for Pt group metals [66]. Besides potential water scarcities, other factors such as high greenhouse gas (GHG) emissions, extensive land transformation, and toxification are crucial factors that are for example endangering living space, biodiversity and agricultural land [66]. LCA, which is implemented in well-defined ISO standards [67,68], quantifies and evaluates such environmental impacts [69]. A description of the major steps in the LCA methodology can be found in the SI note N2 and in references [70,71].

Table 1

Required precursor masses for 1 kg perovskite. The molar content of 1 kg perovskite was calculated for $La_{0.6}Sr_{0.4}CoO_3$ (4.4 mol) and $La_{0.6}Ca_{0.4}FeO_3$ (4.9 mol). The required stoichiometric amounts for each cation were calculated accordingly and related to the required mass of the precursor material. The oxygen content was not stoichiometrically balanced here, as it originates from precursor anions such as the nitrates or acetates in the later stage of fabrication as well as from oxygen in the air in the calcination step.

Perovskite precursor	Precursor mass required for 1 kg perovskite [g]
$La_{0.6}Sr_{0.4}CoO_3$	
La_2O_3	434
$SrCO_3$	262
Co_3O_4	356
$La_{0.6}Ca_{0.4}FeO_3$	
La_2O_3	481
$CaCO_3$	197
Fe	275

In this paper, we identified the environmental impacts of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ precursors serving as catalysts at the anode of AWE based on a LCA approach. The system boundary for the LCA covers the entire precursor fabrication process including raw material extraction and processing with all necessary transportation steps (see also Fig. 4).

The environmental impacts were conducted with the environmental footprint method (EF) 3.1 that is recommended by the European Commission [70,71]. The EF methods were developed by the European Commission in 2013 and updated in 2021 (EF 3.1) [72,73]. The EF methods include the Product Environmental Footprint (PEF) and the Organization Environmental Footprint (OEF). They provide detailed guidelines for modeling, calculating and reporting the environmental impacts of products and organizations over their entire life cycle, based on established international practices and standards such as ISO 14040/44 [67,68]. This fosters transparency, provides access to credible environmental information and supports informed decision-making by companies and consumers, making it a crucial enabler on the path towards a more sustainable economy. We choose the so-called midpoint category approach (and not endpoint approach), because it differentiates environmental impacts in more detail, facilitating identifying possible environmental hotspots. Further, they are easier to compare to other studies and they are less susceptible to uncertainties as they are closely linked to the LCA inventory data [74]. The impact categories evaluated here are climate change, water use, land use, acidification, human ecotoxicity, freshwater ecotoxicity, resource use - minerals and metals, eutrophication marine and freshwater, ozone depletion, photochemical ozone formation and ionizing radiation. Table 2 shows the impact category indicators and units of the evaluated categories.

Further, we determined the so-called “functional unit” (FU) that is important for an appropriate comparison of different products or techniques by LCA. In this study, 1 kg catalyst load of the two perovskites serves as FU because both perovskites show comparable catalytic mass activity and hence, produce comparable amounts of oxygen (and hydrogen) gas. As detailed in section 3.1, the mass activity was determined by CV. This FU is a simplified approach compared to FUs used in LCAs of more mature hydrogen technologies, where typically the catalyst mass per kW or per kg H_2 over the lifespan of the electrolyzer is chosen [75]. However, the choice of mass as FU is useful for comparing initial impact differences and identifying possible hotspots of the precursor materials. Here, the FU is hence chosen based on the initial activity obtained in rotating disk electrode (RDE) studies, but has to

Table 2
Considered impact categories and the corresponding units [70,71].

Impact category	Explanation and unit
climate change	global warming potential in kg CO_2 equivalents (eq.)
water use	deprivation weighted water consumption in m^3 world equivalents (eq.)
land use	soil quality index in points (Pt)
acidification potential	accumulated exceedance of acids in the exposed area in mol of H^+ eq.
human toxicity, cancer	comparative toxic unit for humans in CTU_h
ecotoxicity, freshwater	comparative toxic unit for ecosystems in CTU_e (comparative toxic unit for aquatic ecotoxicity impacts)
Resource use - minerals and metals	abiotic resource use of minerals and metals in kg antimony (Sb) eq.
eutrophication, freshwater	fraction of nutrients reaching fresh water endcompartment in kg P eq. (impacts of nutrients relative to P phosphorous containing nutrients)
eutrophication, marine	fraction of nutrients reaching marine end compartment in kg N eq. (impacts of nutrients relative to N nitrogen containing nutrients)
ionizing radiation human health	human exposure efficiency relative to U235 in kBq U235 eq. (Becquerel)
ozone depletion	ozone depletion potential in kg CFC-11 eq. (chlorofluorocarbon-11)
photochemical ozone formation, human health	tropospheric ozone concentration increase in kg NMVOC eq. (non-methane volatile organic compounds)

neglect aspects of load optimization, integration and final stack lifetime. A similar approach was chosen by Mori et al. for their hydrogen technology LCA study where the FU is also set to the mass of the relevant material [76]. Therefore, recurring analysis of sustainability parameters and a respective refinement of the FU are crucial at higher level of integration and technology.

The environmental impacts were taken from the ecoinvent3.9 database, based on the precursor masses in Table 1. The datasets used for the different precursors can be found in the SI Table T2. These aggregated data represent the global production of the material precursors and include all processes from raw material extraction to energy supply, transport, and processing. The aggregated data is typically used in LCAs for background processes.

If the technical performance is significantly different, the required material mass can be scaled (via e.g. a larger geometric surface area) to achieve similar hydrogen outcomes. This automatically leads to a higher material consumption for the catalyst with smaller mass activity [77].

2.4. Social parameters

The social impact of the energy transition is underexplored. And it is still an open question which materials can bring the biggest social benefits but also pose risks [78]. Energy material supply chains are recently intensively discussed regarding their social impact especially on workers and local communities of raw material extraction [21,79]. For those cases, the UN has published human right guidelines for enterprises and governments [80] (implemented in the social responsibility standard SA8000), tracking child labor, forced work, health and safety, discrimination of employees and much more [81,82]. Moreover, the EU just recently passed the law for social responsibility throughout the whole supply chain [83]. For energy materials such as noble metals or transition metals the often-mentioned hazardous conditions in artisanal mining are criticized [79]. The hazardous working conditions are even more dramatic when child labor is involved [21].

Qualitative and (semi-)quantitative tools for a social life cycle assessment (sLCA) have been developed [84,85] and applied to energy technologies [86–88]. However, social impact assessment methodologies have a lower stage of maturity than environmental LCA [85]. Typically, sLCA requires extensive data collection, often inaccessible on the level of early-stage research. Hence, this study considers existing literature for a qualitative identification of hazardous working conditions and the existence of child labor for the raw material extraction of the precursors for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$.

2.5. Material recycling

Recycling and substitution of critical materials are risk minimizing factors as they directly reduce the dependency of the raw material supplying countries and companies [12,89]. This is reflected in the calculation of the supply risk in the EC-CRM 2023 [12]. However, today's recycling rates and secondary material usage are relatively low for many relevant materials [12,89]. For example, the “End-of-life Recycling Input Rate” (EOL-RIR) [89] of the platinum group metals and the rare earth metals is only 12 % and 1%, respectively [12]. In this paper, the current EOL-RIR was considered as a separate indicator to identify if sufficient secondary material supply exists for the relevant precursor materials of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$. Then, we assess if the available recycled materials are suitable for the projected perovskite synthesis. Based on this, recycled precursor materials are considered for the sustainability evaluation.

2.6. Technical performance

In early-stage energy materials research, the focus typically lies on the understanding of physical and chemical structure-property-relations to reveal new material design rules that can lead to higher application performances. The material composition, efficiency and durability can improve the sustainability of the technology's whole life cycle, as the economic, environmental, and social impacts directly scale with the required material consumption.

On the one hand, early-stage materials research is often based on model systems that depart from the real geometry in the final application, leaving significant room for improvements by engineering of the final assembly. On the other hand, such model systems have the advantage to facilitate a systematic chemical and physical analysis of the materials of interest and at the same time allow a precise comparison of a broad range of materials. In this paper, we compare the OER catalytic mass activity of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ thin films that have the advantage of providing comparable surface morphologies facilitating the observation of intrinsic catalytic properties.

3. Case study: $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ catalysts in alkaline water electrolysis

3.1. Catalytic activity results of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ thin films

To compare the intrinsic catalytic activity of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ in the OER, we deposited 20 nm thick catalyst layers by pulsed laser deposition to achieve comparable surface morphologies that have a route mean square roughness of 3-5 nm (see atomic force microscopy scans in the SI Figure S1a). For both samples, the exposed catalyst surface area was similar to the geometric surface area, with deviations of less than 4 %. Therefore, the small surface roughness differences have no impact on the OER activity (SI Figure S1a). A comparable surface morphology facilitates obtaining the intrinsic catalytic activity decoupled from larger surface area differences typically appearing with powder synthesis routes [52]. Powder catalysts in commercial electrolyzers exhibit orders of magnitude higher surface area. This translates into manifold higher currents for the same electrode area (electrode footprint) compared to the investigated thin film catalysts [90] which is important to consider at a higher integration level of the catalysts. The crystallographic characterization of the thin film stack can be found in the SI Figure S2. A platinized Si substrate was chosen as current collector to avoid contact resistances (see sample stack in

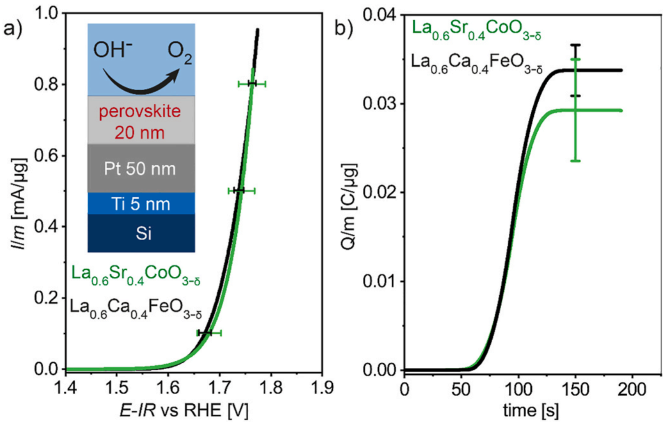


Fig. 3. a) OER catalytic mass activity is shown from a CV scan of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ thin films. Anodic and cathodic scans were averaged to remove capacitive current contributions. b) Charge per µg catalyst passed in one CV cycle. A comparable charge is generated by both perovskites within one CV scan.

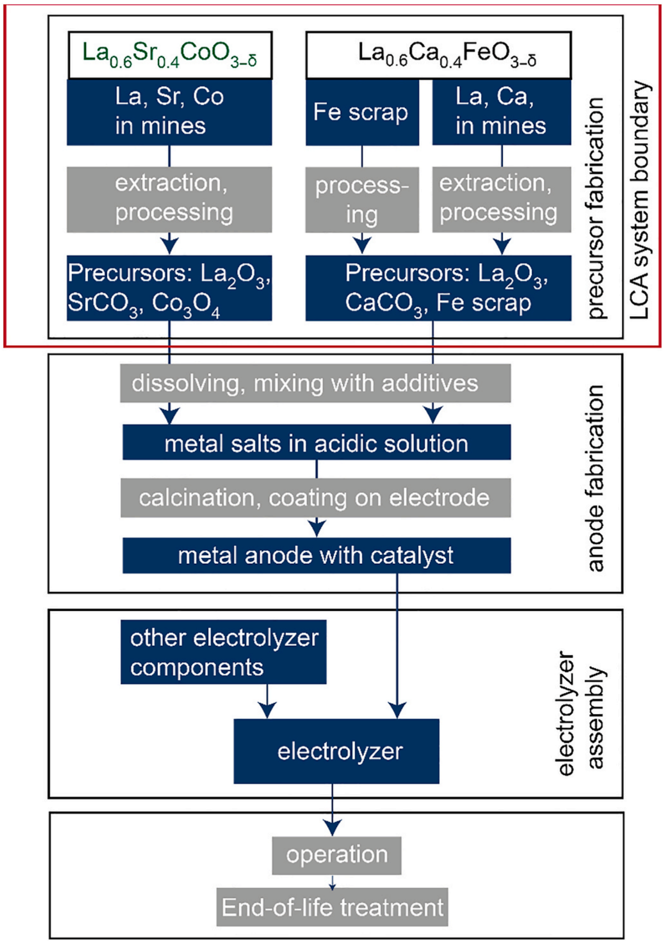


Fig. 4. Life cycle of an alkaline water electrolyzer with projected production pathways for perovskite oxides $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ catalysts for the anode. The LCA system boundary includes material extraction, transportation, and precursor production.

Fig. 3a; 5 nm Ti were used as adhesive interlayer). Note that the Pt layer itself has a much lower OER activity than perovskite catalysts [91] (SI Figure S1d), so that it serves merely as electrical contact and does not contribute to the intrinsic activities of our model perovskites. Moreover, the Pt layer is fully buried underneath the perovskite layer as demonstrated by x-ray photoelectron spectroscopy measurements presented in the SI Figure S1e, so that any contribution of the Pt layer to the observed OER activity can be excluded.

To relate the OER intrinsic activity to the catalyst mass loading (i.e., to determine the mass activity), CV was conducted. We determined the OER activity of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ perovskite oxide thin films in a RDE setup in a 3-electrode configuration. Fig. 3a shows representative averaged CV scans obtained for the two thin films. Both perovskites exhibit comparable OER mass activity reaching about 0.75 mA/µg at an anode voltage of 1.76 V vs RHE (reversible hydrogen electrode). The standard deviation of the CV potential is 9-11 mV for $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ and 20-25 mV for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and is calculated from three different samples of each perovskite. It was determined at three different currents along the CV scan at 0.1, 0.5 and 0.8 mA/µg (the standard deviation differences of 9-11 mV and 20-25 mV stem from these three different points).

As shown in Fig. 3b, the total charge converted during the CV scan per µg catalyst is comparable for both catalysts, with a slightly larger value obtained for $La_{0.6}Ca_{0.4}FeO_{3-\delta}$. Hence, a similar amount of oxygen and respectively hydrogen were produced, implying a comparable performance of both materials for their envisioned application as OER catalysts. Although the chemical composition of the perovskite varies on the A-and B-site, the intrinsic catalytic activity is comparable. This is in contrast to some studies where ferrates, and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ in particular [51], are sometimes linked to a low OER activity compared to cobaltates [92,93]. However, as we could show in a previous study [94], $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ has a high intrinsic catalytic activity comparable with the highly active and well-studied $LaNiO_{3-\delta}$ and $La_{0.6}Sr_{0.4}CoO_{3-\delta}$, which is evident when the $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ is properly electrically contacted, as in our case with the underlying metallic current collector.

While these lab-based RDE measurements are ideal for determining relative catalytic activity, they are less ideal for the determination of the catalyst stability in a final application scenario [95,96]. Particularly, degradation rate and lifetime depend on the system integration of the catalyst layers; enhanced lifetimes were reported for integrated catalysts in membrane-electrode assemblies compared to RDE experiments [95,96]. While these studies confirmed that accurate stability tests under industrial conditions are inaccessible using a model-system approach and testing setup, various model-system studies of these and similar compounds have shown that very thin layers of perovskite oxides are sufficiently stable to serve as model systems to assess and compare activity trends [33,97,98]. We note that similar perovskite catalysts have been successfully subjected to degradation studies under 1000 mA/cm^2 on a highly integrated level [90], highlighting their suitability for application. Regarding the role of the underlying Pt layer, we observed about 60x greater OER activity for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ compared to Pt, implying that the measured OER currents cannot result from exposure of the underlying Pt layer (Fig. S1d in the SI). We note that final stability benchmarks have to be established in a recurring manner at higher integration levels to yield additional and more precise input parameters for the sustainability evaluation towards industrial levels. Here, we rely on the intrinsic activity comparison as a starting point for a comprehensive material evaluation in early-stage catalyst research. The similar mass activity and generated charge in one CV cycle suggests that comparable amounts of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ catalysts may be projected to achieve similarly efficient hydrogen production in the final electrolyzer assembly. We hence elaborate economic and environmental aspects based on the FU of 1 kg perovskite.

3.2. Projected synthesis routes of perovskites on large area electrodes

While the determination of the intrinsic catalytic activity of the selected materials is done ideally on laboratory- and model-system level, a quantification of the projected sustainability parameters must consider the projected industrial-scale production route. Perovskite oxides for industrial applications are typically synthesized via solid-state or wet-synthesis methods [39,99]. Since the solid-state route is more susceptible to impurities, requires higher reaction temperatures, and typically forms larger particles which is coupled to a lower specific surface area [39,99], we consider the wet chemical methods to achieve high-purity nanoparticles for a potential industrial perovskite oxide synthesis. Note that identified precursors are also suitable for the solid-state route besides Fe scrap as that would have to be converted to iron oxide first [100].

Fig. 4 illustrates the life cycle of an alkaline water electrolyzer with projected synthesis routes for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$, starting from the raw material input to the finally coated electrode for AWE. In this scenario, La, Sr, and Co containing rocks are extracted and further processed to La_2O_3, $SrCO_3$, and Co_3O_4. For the $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ production La and Ca containing rocks are extracted from mines and processed to La_2O_3 and $CaCO_3$. In contrast, Fe can be used from iron scrap, since high amounts of iron scrap are available on the scrap market. The iron scrap can be directly converted to iron nitrate [101] or another organic metal salt, and hence generally bypass the mining process. Note that not all scrap types are applicable. Here, an alloy free [102] and new iron scrap can be chosen and potentially be used for the perovskite oxide synthesis. More details about the option to use recycled materials can be found in the material recycling section below.

In the subsequent production step, the precursor materials are dissolved in acidic solution to form metal nitrates or other metal-organic acid salts. Stoichiometric amounts of the precursor solutions are then mixed, combined with additives and thermally treated to arrive at fine nanoparticle perovskite powders. Different synthesis options for this have recently be considered in Ref. [103]. The nanoparticles are then deposited on large-area metal electrodes via for example screen printing [104,105], jet printing, tape casting, or electrochemical deposition (Fig. 4 bottom) [39,100,106].

The latter process steps to arrive at the functional nanoparticles are typically quite similar for the perovskite family, and $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ in particular as seen in Refs. [107,108]. Therefore, for a direct comparison of the two perovskites, we can focus on the production of the precursor materials La_2O_3, $SrCO_3$, $CaCO_3$, Co_3O_4 and iron scrap (marked in Fig. 4 with the box "precursor fabrication"). The main goal is to identify if there are material hotspots in the steps of precursor extraction and processing by analyzing the aggregated data of the ecoinvent database.

The subsequent common process steps will similarly add to the full sustainability evaluation, e.g. through the calcination step which requires a high energy input but can be regarded similar for both materials under test. In the following, the material criticality, raw material prices, as well as environmental and social impacts resulting from the precursor masses for 1 kg perovskite (see Table 1) are discussed with a focus on the precursor materials.

3.3. Sustainability evaluation results

3.3.1. Material criticality

In the current EC-CRM 2023 assessment, a material is considered critical when the supply risk (SR) parameter exceeds a value of 1 and the economic importance (EI) parameter exceeds a value of 2.8 (shaded area in Fig. 5) [12,28]. As shown, Sr, Co and La are hence considered as critical in the 2023 EU assessment whereas limestone (the major source for Ca) and Fe ore are considered as non-critical. Both, limestone and Fe ore have a high economic importance for the EU, but the supply risk is low because of a high diversification level of supplying countries inside and outside the EU [12].

Sr is evaluated as critical, primarily because it is essential to produce magnets and pyrotechnics and has only one EU supplying country (Spain). It is hence affected by both a high economic importance as well as a high supply risk. Co is evaluated as critical primarily because its supply is poorly diversified, and the Democratic Republic of Congo (with 63 % the main global Co supplier) is suffering from high political instability. Furthermore, Co is essential to produce several alloys and hard materials that add high value to the EU economy. La is critical due to a high supply risk of 3.5 and a high economic importance of 2.9. This is because 85 % of the global La supply is coming from China where La is extracted as well as processed. Since La is not recycled and not easily substituted in relevant applications, the EU relies to a large extent on La import from China. La mainly adds value to the EU economy as component in fluid cracking catalysts and other catalysts [12]. While the depletion of known La reserves is projected to occur within the next 100-500 years [49], the high geopolitical dependencies and low supply chain diversification for La supply can be a significant supply risk in the closer future.

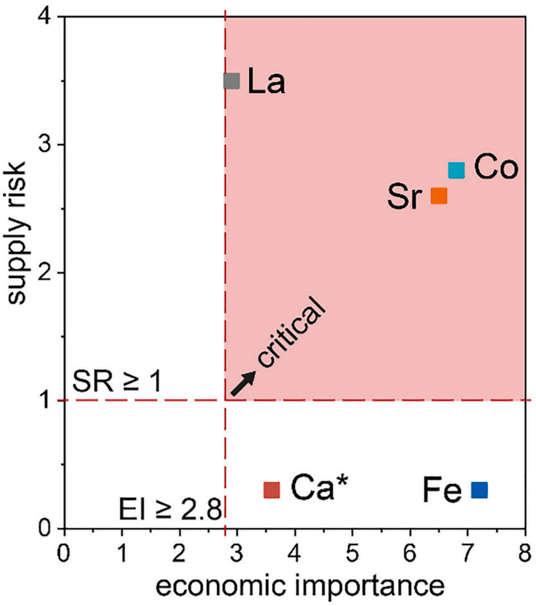


Fig. 5. EU criticality assessment for the raw materials used for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ adapted from Ref. [12]. Ca* is limestone and Fe the iron ore [12].

Based on this evaluation, exchanging Sr and Co for Ca and Fe can reduce the overall material criticality of our model catalysts. However, La is of immediate concern for both perovskites regarding its high supply risk.

Projected increase in demand for perovskite oxides in the EU based on experimental results

To estimate how many tons of perovskite would be required for AWE in a gigawatt scale in the EU in the future, we calculated the power-to-mass ratio based on our model systems and compared it to an estimated hydrogen demand scenario in 2050 based on Ref. [109]. With that, it can be estimated if some of the perovskite precursors would require larger supply chain expansions than others in comparison to today's EU material consumption.

Here, we calculated the mass-to-power ratio at 1.76 V vs RHE with an output of 0.75 mA/µg. At this operating voltage the required perovskite mass-to-power ratio is around 0.76 t/GW under the assumption that the perovskite catalysts form dense 20 nm thick layers on commercial electrode surfaces and are operated in 0.1 M KOH solution at room temperature (as according to our experiments). Note that AWE electrolyzers are typically operated in 5-10 M KOH solution at 40-85 °C [8,110,111]. Hence, the industrial operating conditions have an influence on the final mass-to-power ratio of the perovskites as well as the operational catalyst lifetimes. However, this calculated mass-to-power ratio is comparable to the estimate of Ir catalyst ratio which is 0.75 t/GW in acidic proton exchange membrane electrolysis [30]. The comparability of catalyst loads in AWE is hard because Ni as the main catalyst material is also applied as substrate and as bipolar plates material [30].

To upscale the power-to-mass ratio to the forecasted EU hydrogen consumption in 2050 it is estimated that 429 GW of AWE power is required in a high demand scenario (HDS) (calculations found in SI table T3 [109]). To cover this demand the perovskite catalyst layers investigated here would sum up to around 325 t perovskite. As not all AWE plants will be operated with perovskite oxides, this number is an upper limit for the experimental results. Such perovskite catalyst loads in AWE correspond to 5-6 % of the recent, annual EU lanthanum consumption [112] and 0.8 % of the average EU Co consumption [113], whereas the other elements Sr, Ca and Fe fall below the average EU consumption of 0.1 % [114–116] (see for details of the EU material consumption SI table T3).

As we chose thin film catalysts that have comparably smooth surface morphologies to obtain a fair comparison of the catalytic performance, final mass-to-power ratio may be further optimized and influenced by material morphology engineering, catalyst integration, and by the application of industrial operation conditions. However, an estimate of possible material consumption gives an overview of possible supply bottle necks. The relatively larger supply chain expansion of La compared to the other precursors as well as lanthanum's high material criticality might hence lead to a threat for the application of both perovskites. All rare earth elements are classified as critical raw materials in the EC-CRM [12] report and their demand in the energy sector is increasing sharply [3]. Hence, the rather monopolistic supply chain of rare earth elements is alarming for the energy transition.

3.3.2. *Economic impact based on precursor prices*

The market prices of the required precursor materials reflect a direct indicator of the economic impacts in the materials selection. Fig. 6 shows the cumulative precursor price per kg perovskite for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$. Detailed price information and sources are summarized in SI Table T3. The cumulative precursor price for 1 kg perovskite is about 10 times lower for $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ compared to $La_{0.6}Sr_{0.4}CoO_{3-\delta}$. For $La_{0.6}Sr_{0.4}CoO_{3-\delta}$, the cumulative price is dominated by Co_3O_4 while La_2O_3 and $SrCO_3$ share only ~10 % of the precursor costs. For $La_{0.6}Ca_{0.4}FeO_{3-\delta}$, the iron substitution reduces the cumulative price by a factor of 10, where iron scrap makes up only 15 % of the projected $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ price. The rest of the cumulative price is shared by La_2O_3 and $CaCO_3$ with 57 % and 27 %, respectively.

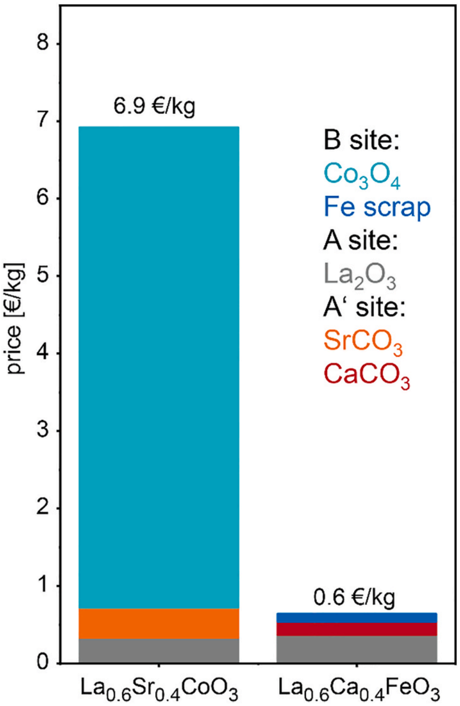


Fig. 6. Cumulative precursor prices for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ per kg perovskite.

3.3.3. *Environmental impacts of precursor production*

Fig. 7 shows the environmental impact results of the precursor materials for 1 kg perovskite oxide for the categories climate change, water use, land use, acidification potential, human toxicity and ecotoxicity. Further impact category results on resource use - mineral and metals, eutrophication, photochemical ozone formation, ozone depletion and ionizing radiation are shown in the SI Figure S3. Detailed definitions of the single impact categories can be found in Ref. [71].

For all environmental impact categories from Fig. 7 and Fig. S3, the use of Fe from scrap instead of Co from Co_3O_4 for the perovskite oxide B-site significantly decreases the environmental impact, particularly for the categories climate change, water use, acidification potential, and human toxicity. In contrast, $SrCO_3$ and $CaCO_3$ precursors both have almost no environmental impact in all categories. For $La_{0.6}Ca_{0.4}FeO_{3-\delta}$, the La_2O_3 production dominates in all impact categories by at least 90 %. Also, for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$, the La_2O_3 production makes up at least 75 % of the environmental impact in the four categories land use, ecotoxicity freshwater, marine eutrophication, and ozone depletion, indicating that the use of La in both catalysts implies severe environmental burdens. More detailed results about the emissions which cause the environmental impact can be found in the SI (Fig. S3).

As discussed in section 3.2, different perovskite nanoparticle synthesis routes are possible, where both perovskites can be produced under similar conditions as reported elsewhere [107,108]. The further processing of the precursor materials to the final perovskite nanoparticles will add up to the overall assessment, however in a similar manner for both. Nandikes et al. recently published an LCA study on various $LaFeO_3$ nanoparticle synthesis routes [103] and consistently found that the La precursor production is the predominant factor in several impact categories. Therefore, decreasing the La amount in the perovskite would support the overall environmental footprint of the nanoparticle synthesis. Another LCA study referring to high temperature solid oxide electrolyzers with a $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ air electrode, reported a higher global warming potential per kg of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ compared to our study [117]. Yet, this difference stems presumably from the final synthesis step requiring sintering and firing at high temperatures. However, for perovskites used in low temperature AWE, nanoparticle synthesis routes such as suggested by Nandikes et al. would be more suitable and require less extreme temperatures.

3.3.4. *Social aspects*

A quantitative analysis of the social impacts is elusive on the level of early-stage research. However, we highlight some key aspects that can yield a preliminary assessment of the expected social impacts of raw material extraction.

With respect to the Co mining industry, social impact assessments exist mainly for the Democratic Republic of the Congo, as main supplier of Co. While there are regulated working standards for artisanal mining in the Democratic Republic of the Congo, many mining sites do not follow those rules and many artisanal miners do not have the knowledge about these regulations. Hence, mining tunnels are exceeding the allowed tunnel length, and several safety constructions are often not installed [118]. Although child labor is also regulated in the Democratic Republic of the Congo, 29 % of the miners were children in 2011, reflecting the high poverty in the country [118]. In contrast, the use of iron scrap is generally acknowledged to have a positive social impact as, for example, the use of iron scrap reduces climate change related socio-economic costs globally, as it reduces CO_2 pollution [119].

The assessment of social impacts in the La industry is less mature [120]. However, it is reported that the rare earth industry in China

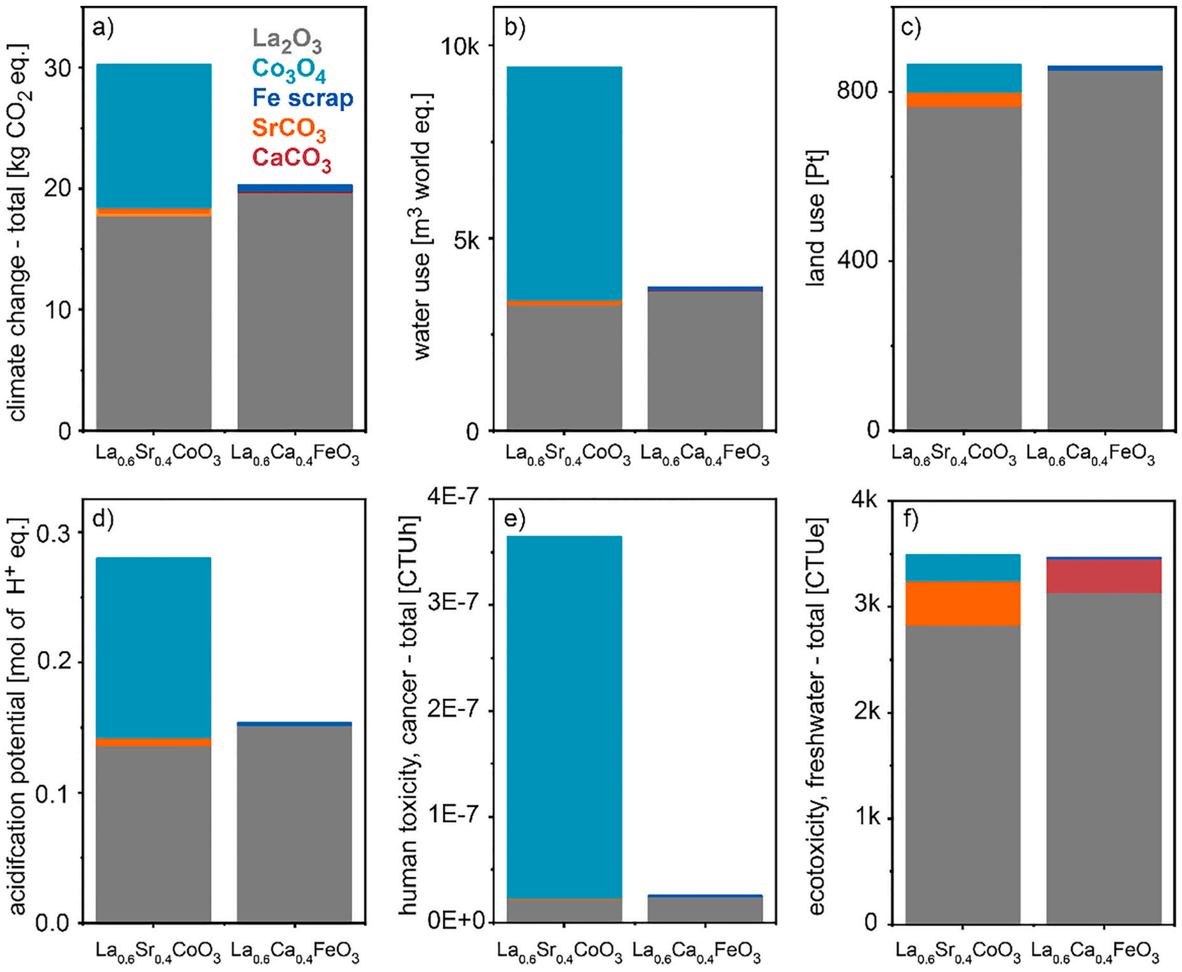


Fig. 7. Environmental impact categories: climate change, water use, land use, resource use mineral and metal, acidification potential, human and freshwater ecotoxicity of the precursors required to produce 1 kg of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$.

causes health problems for the directly involved stakeholders such as increased exposure to radioactive tailings [120–124]. Moreover, it is stated that the environmental burdens of rare earth material processing are so high that it is hard to get a permission for rare earth processing sites in developed countries [120]. Therefore, rare earth material processing takes place mainly in less developed countries [120].

In contrast, with Spain as a major supplier for Sr in the European Union [115], child labor and hazardous working conditions are regulated and can be considered as negligible in the European Union. Likewise, Ca is also locally sourced in Europe [116] and to our knowledge no child labor or hazardous working conditions are reported.

3.3.5. *Material recycling*

Environmental, economic, and social impacts as well as the material criticality, can be potentially improved if recycling rates increase and accessible secondary material supply is provided, especially in the case of La and Co, as critical raw materials with high environmental impact in the precursor production.

Today, the EOL-RIR of Co is already around 22 % [12]. However, the recycled Co is to a large extent embedded in Co-alloys and therefore not directly accessible for Co_3O_4 recovery [125]. The use of recycled Co from battery cathodes would potentially be suitable for the perovskite production, but competes with the battery market itself [126,127]. Therefore, our case study is based on the primary (non-recycled) material supply for Co_3O_4.

In contrast, Fe has a high recycling rate (EOL-RIR 31 % [12]) and certain scraps are suitable for the precursor production. Fe scrap can be domestically available and Fe scrap contributes to the comparably low Fe market prices and the lowered environmental and social impacts.

The EOL-RIR of La is only 1 % [12], and hence, for the perovskite production not applicable at this stage. The same holds for limestone with an EOL-RIR <1 % [12]. However, as Ca is a non-critical material and $CaCO_3$ production from primary sources has a low environmental impact, the need for increased recycling rates is of lower priority. The Sr EOL-RIR is nearly zero [12] and hence not considerable for secondary material input today. Given the material criticality of Sr, Co and La, efficient recycling may reflect a future strategy to mediate the high supply risk for the primary material supply.

4. Discussion

All analyzed sustainability parameters of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ OER catalysts are summarized and compared in Table 3. While the two perovskites exhibit a comparable technical performance in our laboratory-based experiments, several sustainability parameters of $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ have a lower impact compared to $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ (marked in green/red color in Table 3). This includes that the supply risk of Ca and Fe is low compared to Sr and Co. Moreover, the projected price of $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ is 90 % lower than for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ because Co is avoided. The environmental impact of $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ is at least 25 % lower compared to $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ in those categories, where cobalt oxide production is having a significant impact. However, in the environmental impact categories that are dominated by La_2O_3 production, both $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and

Table 3

Summary of the sustainability parameters of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ as alkaline oxygen evolution catalysts.

sustainability parameters	**$La_{0.6}Sr_{0.4}CoO_3$**	**$La_{0.6}Ca_{0.4}FeO_3$**
material criticality (EC-CRM 2023)		
supply risk (≥ 2.8 🔴)	La 🔴 Sr 🔴 Co 🔴	La 🔴 Ca 🟢 Fe 🟢
economic importance (≥ 1 🔴)	La 🔴 Sr 🔴 Co 🔴	La 🔴 Ca 🔴 Fe 🔴
economical impact		
cumulative precursor price	🔴	🟢 90 % lower
environmental impact (LCA EF 3.1 ecoinvent 3.9) similar impact ⚫ | at least 25 % higher 🔴 | at least 25 % lower 🟢		
climate change (CO_2 equivalents)	🔴	🟢
land use	⚫	⚫
water use	🔴	🟢
acidification·	🔴	🟢
human toxicity	🔴	🟢
ecotoxicity	⚫	⚫
ozone depletion	⚫	⚫
photochemical ozone formation	🔴	🟢
eutrophication (freshwater)	🔴	🟢
eutrophication (marine)	⚫	⚫
ionizing irradiation	⚫	⚫
social impact **yes 🔴 no 🟢**		
child labor	La not known, Sr 🟢 Co 🔴	La not known, Ca 🟢 Fe 🟢
hazardous working conditions	La 🔴 Sr 🟢 Co 🔴	La 🔴 Ca 🟢 Fe 🟢

$La_{0.6}Ca_{0.4}FeO_{3-\delta}$ have similarly high environmental impact (marked in black). $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ has potentially a lower social impact than $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ because Co mining often involves child labor and can occur under hazardous working conditions. Nevertheless, La processing can also occur under hazardous working conditions which is a disadvantage for both.

In summary, the exchange of Co to Fe may hence be assessed as generally beneficial for the majority of sustainability parameters. Similarly, the exchange of Sr by Ca further reduces the overall supply risk of the perovskite. Nevertheless, the presence of La yields a significant challenge for the catalyst usage in larger scales for both perovskites and has to be addressed in the future.

In agreement with these points, the EU 2023 foresight study shows that La, Sr or Co are ranked at the top of the different possible supply risks for an AWE stack [103] whereas commercial alkaline water electrolyzers do not rely on these raw materials today [109]. Regarding the environmental impact of commercialized AWE stacks, Ni - that is used as anode and cathode catalyst (and as Ni mesh) - exhibits high environmental impact in several categories [128]. The usage of perovskite catalysts on the anode instead of Ni can therefore decrease or increase the environmental impact and raw material supply risk of the AWE stack depending on their final composition, mass load and technical performance.

Our analysis emphasizes that material sustainability evaluation in early-stage research can provide additional criteria for materials selection next to physical and chemical performances [23]. These criteria might act as an additional driver to test and to optimize materials that have a lower supply risk as well as lower economic, environmental and social impacts in their final application [23,24,26]. If the results of the sustainability evaluation are not as clear as in this case study (a clearly preferred perovskite), multi-criteria decision analysis (MCDA) can be used to structure decision-making processes. MCDA includes mathematical approaches to cluster many individual results into fewer but more manageable results [129].

At the same time, our case study revealed both the chances and limitations as well as required assumptions to arrive at a quantitative sustainability evaluation. Model catalysts, as employed in this study, are ideal for determining intrinsic catalytic activity as they have comparable surface morphologies and are independent of additional stack assembly related resistances. Hence, for the comparison of two materials that are supposed to serve the same function (here the anode electrocatalysts in AWE), the model catalysts can be quantitatively related to sustainability parameters via their mass activity. However, the scaling to industrial applications is limited, as for example lifetimes and loadings of final electrolyzer stacks are hard to predict which is indispensable to project and approximate the final electrolyzer production and operation impact. Importantly, the FU of 1 kg perovskite oxide chosen here delivers a fair comparison in such an early stage of research, while in industrial electrocatalyst research the LCA can be conducted with more commonly used FUs [75]. This enables a comparison to other commercial or prototype AW electrolyzers where realistic lifetimes are considered, catalyst loads are optimized, and the surface roughness is optimized.

Nevertheless, a first evaluation can be made with the model system approach if the assumptions and conditions are well-defined. This can be the basis for material choices in later stages of materials research.

Testing a broad range of materials with catalyst model systems in early-stage research can provide a general basis for preselecting materials in the applied research of energy materials. Implementing a sustainability evaluation already at early stages of materials research can be a strong opportunity to identify less sustainable material compositions and potentially refocus the research. As found in this study, reducing the La content or even eliminating it could improve the sustainability of the material even more as compared to the sole exchange of Co and Sr to Fe and Ca. However, the replacement of La has to be tested for OER catalyst performance to make a further decision on the necessity of La containing perovskite catalysts. One option could be to replace La fully with Ca on the A-site where some recent studies show that $CaCoO_3$ [130] or $CaFeO_3$ [51] show high catalytic activity. Nonetheless, it was also reported that the usage of only alkaline earth elements on the A-site might lead to a faster structural transformation [47,48]. Another option could be to replace La by other rare earth elements such as dysprosium or neodymium but these are also critical raw materials [12] and do not have a lower environmental impact [131,132]. Hence, they are not necessarily suitable for a La exchange.

There is a recent and important study [18] that tests several lanthanide substituted $RBaCuFeO_{5-\delta}$ (with R = lanthanides) perovskite oxides OER catalysts. The motivation of that study was to avoid Co based perovskites, as Co is suffering from high prices and is competing in the battery market. This approach is crucial for a holistic material choice, yet, considering the e.g. material criticality of the investigated lanthanides can further improve it. This is an example where the materials science community can benefit from knowledge on e.g. precursor environmental footprints and criticality assessments to tackle holistic material choices from the onset.

In further feedback loops, the sustainability of energy materials can be evaluated both in applied research (e.g., testing the catalyst in an actual electrolyzer stack) as well as in industrial research (e.g., long term tests under industrial conditions) (Fig. 8). The latter includes the development of a sustainable material design strategy for final scaling parameters and long-term technical performance. Such a feedback loop between energy materials research and sustainability evaluation would be beneficial at all stages of research to constantly reevaluate and leverage the sustainability of material-intensive technologies.

The factors discussed here, such as the use of critical raw materials, precursor price distribution, and the ecological and social footprint of preliminary products (here precursors), also remain relevant in applied and industrial research. There they are incorporated into social, economic and ecological life cycle assessments over the entire product life cycle, which are already established today.

Many industrial companies, including those in the energy sector, are feeling increasing economic pressure and are facing increasing regulatory requirements to reduce their environmental footprint and to avoid possible supply bottlenecks in times of tensioned geopolitics [133]. Thus, material scientists can consider and even influence the design of materials in the early development phase that meet these needs from the onset. Such evaluation can yield an initial guidance for expected sustainability bottlenecks also at a higher level of integration and technology.

5. Conclusion

We showcased to which extent a sustainability evaluation can be conducted for model OER catalysts in early-stage research. First, material criticality assessments that include raw material economic importance and supply risks in the context of geopolitical constraints are a more powerful indicator of possible supply chain bottlenecks than mere 'earth abundance' maps that are solely geological perspectives. Furthermore, based on projected industrial synthesis routes, economic and environmental parameters of precursor materials can already be related to the mass activity of model catalysts in the lab-scale. Environmental impact categories considered in LCA can be assigned to the electrocatalyst precursor materials uncovering possible hotspots. Recurring LCAs at higher technology integration levels enable realistic electrocatalyst stability tests, which is out of the scope in such early stages. Social impacts can also be considered on a qualitative scale to identify if, for example, poor working conditions exist in raw material extraction. Moreover, secondary material input from recycling presents an attractive parameter if sufficient supply is offered on the commodity market.

Based on such analysis, material hotspots can be identified, and new material design strategies can be established already in early stages of energy materials research. Striking a proper balance between sustainability and technical performance in constant feedback loops at all stages of energy materials research might benefit the energy transition.

Our case study shows that in the future such sustainability evaluations in early-stage materials research can be enhanced and facilitated through a broader dissemination of critical raw material assessments such as the EC-CRM report, an overview of the environmental footprints of common precursors, and enhanced cooperation options between materials scientists and sustainability as well as LCA experts. While it remains of utmost importance not to generally restrict the elemental choices in fundamental materials research and to explore unknown

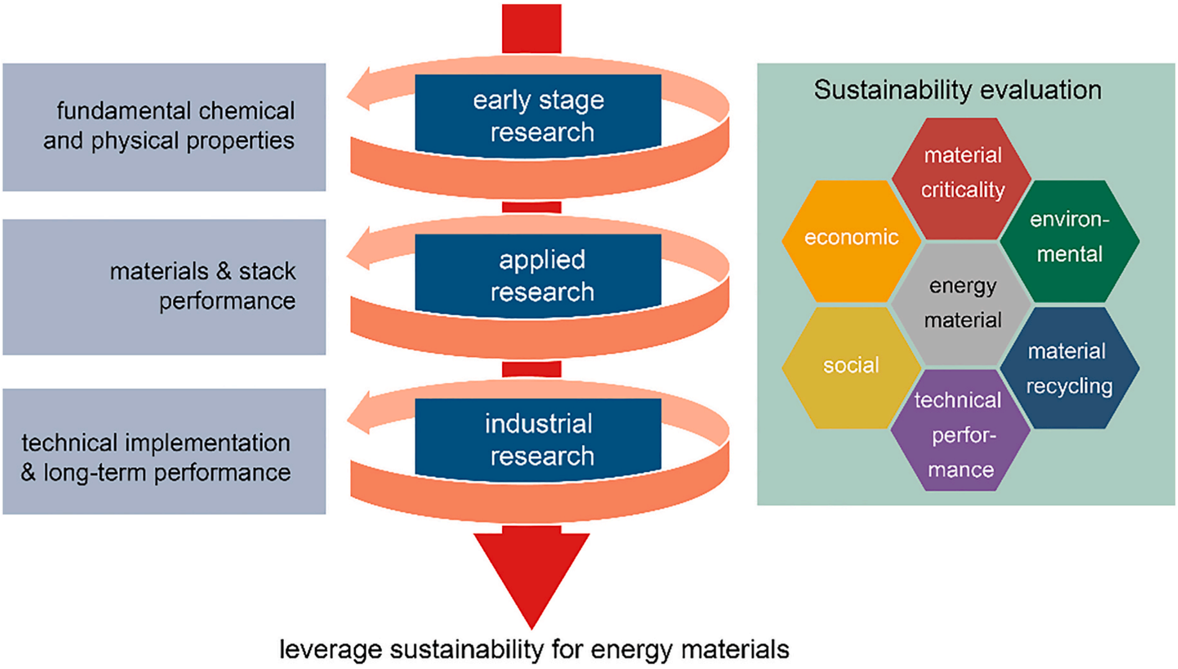


Fig. 8. Approach of holistic material choice for electrolyzers, fuel cells and batteries in early-stage, applied and industrial research.

properties and phenomena independent of industrial scenarios, considering sustainability aspects at an early stage can help to design technologies in a holistic and future-oriented way. As the energy transition requires massive material consumption, and an established hydrogen economy is only one branch of it, early-stage materials research with less critical materials, higher recycling rates, lower environmental, social and economic impact can drive a successful and especially feasible energy transition.

As the energy transition requires massive material consumption, and an established hydrogen economy is only one branch of it, early-stage materials research with less critical materials, higher recycling rates, lower environmental, social and economic impact can drive a successful and especially feasible energy transition.

6. Experimental methods

Thin film fabrication: The thin films of $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ and $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ were fabricated with pulsed laser deposition (TSST, B. V., Netherlands) from ceramic targets provided by Toshima Manufacturing Co., Ltd. (Japan). For the perovskite oxide growth, an oxygen partial pressure of 0.05 mbar, deposition temperature of 650 °C and a laser energy of $2.2\,J/cm^2$ ($La_{0.6}Ca_{0.4}FeO_{3-\delta}$) and $2.6\,J/cm^2$ ($La_{0.6}Sr_{0.4}CoO_{3-\delta}$) of a KrF excimer laser ($\lambda = 248$ nm) were applied. $10 \times 10\ mm^2$ single crystalline silicon substrates (from Siegert Wafer, Germany) were sputtered (Univex sputter tool, SIEMENS, Germany) with a 5 nm Titanium adhesive layer for the subsequent Pt sputter deposition of 50 nm. The platinized silicon substrate was used to ensure metallic contact.

Thin film characterization: The surface morphology was determined with an atomic force microscope (Cypher SPM, Oxford Instruments, Research Asylum, Germany) in the tapping mode. The silicon tips were provided by NanoWorld AG (Switzerland) with a force constant of 42 N/m and a mean width of 30 μm. Grazing incidence x-ray diffraction was conducted with a D8 ADVANCE diffractometer (Bruker AXS GmbH, Karlsruhe, Germany) that is equipped with a Cu anode as K_α x-ray source.

Electrochemical characterization: Electrochemical measurements were conducted ina RDE setup (Pine Research Instrumentation, Inc, USA) at a rotation speed of 1600 rpm in a three-electrode configuration where a Pt coil served as counter electrode and a Hg/HgO electrode (152CHI from CH Instruments, USA) served as reference electrode. The samples were placed in a customized polyether ketone (PEEK) cap on the RDE rotation shaft and sealed towards the electrolyte with an O-ring (diameter of 7.5 mm) so that a defined surface area was exposed to the electrolyte. The electrolyte concentration was 0.1 M KOH made from KOH pellets provided by Sigma Aldrich (99.9 %) and deionized water (Milli-Q, >18.2 MΩ cm). Before the start of the measurement, the electrolyte was purged with oxygen for 30 min and also during the experiment. Double layer capacitance measurements were conducted for $La_{0.6}Sr_{0.4}CoO_{3-\delta}$ from 0.1 to 0.2 V vs Hg/HgO and for $La_{0.6}Ca_{0.4}FeO_{3-\delta}$ from 0.2 to 0.3 V with increasing sweep rates from 10 mV/s to 500 mV/s. CV scans for OER activity determination were conducted between 0.2 V and 1.15 vs Hg/HgO. The CV scan was averaged from the back and forward sweep in the OER potential regime. To transfer the values to the RHE scale we measured the electrode potential between an RHE (HydroFlex, USA) and the Hg/HgO electrode in the electrolyte. The potential scale of the CV scans was *iR* corrected, where the uncompensated resistance was determined by the x-axis offset in the high frequency range of a PEIS measurement (20kHz-0.1 Hz with 20 mV amplitude) at open circuit potential.

CRediT authorship contribution statement

Lisa Heymann: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project administration, Visualization, Writing – original draft. **Andrea Schreiber:** Conceptualization, Data curation, Formal analysis, Methodology, Validation, Writing – review & editing. **Christian Pithan:** Formal analysis. **Christoph Baeumer:** Conceptualization, Formal analysis, Methodology, Supervision, Visualization, Writing – review & editing. **Felix Gunkel:** Conceptualization, Formal analysis, Funding acquisition, Methodology, Supervision, Validation, Visualization, Writing – review & editing.

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.mtsust.2026.101329.

Data availability

Data will be made available on request.

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