Chemical Engineering Journal Advances 16 (2023) 100575

Contents lists available at ScienceDirect

Chemical Engineering Journal Advances

journal homepage: www.sciencedirect.com/journal/chemical-engineering-journal-advances

Paving the green path: A novel approach to material manufacturing using LCA

Marcela Frias Ordoñez [a,b,*], Serena Biella [a,b], Ermelinda Falletta [a,b], Claudia L. Bianchi [a,b]

[a] Department of Chemistry, University of Milan, Via Golgi 19, 20133, Milano, Italy
[b] Consorzio Interuniversitario Nazionale per la Scienza e Tecnologia dei Materiali (INSTM), via Giusti 9, 50121 Florence, Italy

ARTICLE INFO

Keywords:
Synthesis routes
Life cycle assessment
$SrTiO_3$
Photocatalyst
Lab-scale
Cradle-to-gate

ABSTRACT

Perovskite oxides have garnered immense interest across diverse fields owing to their exceptional properties. Among these, $SrTiO_3$ is an excellent photocatalyst under UV light and boasts remarkable thermal stability, rendering it ideal for high-temperature applications. While various synthesis methods exist for $SrTiO_3$, prioritizing environmentally friendly processes, even at the laboratory scale, remains crucial.

This study introduces a pioneering life cycle assessment (LCA) comparing six $SrTiO_3$ synthesis techniques (solid-state reaction, sonochemical, sol–gel, hydrothermal, solvothermal, and molten-salt) to gauge their environmental impacts, pinpoint potential concerns, and offer insights for the future. Interestingly, methods like molten salt and sol–gel display higher pollution tendencies due to significant energy usage, particularly during drying and calcination stages. Conversely, the solid-state reaction and ultrasound-assisted methods emerge as sustainable pathways for crafting environmentally conscious $SrTiO_3$ materials. Significantly, electricity emerges as a pivotal factor influencing environmental impact, urging the exploration of alternatives and the need for strategic decisions. This investigation not only sheds light on the ecological footprint of $SrTiO_3$ synthesis but also underscores the urgency for adopting greener approaches in materials manufacturing.

1. Introduction

Sustainable development was born to balance economic growth, care for the environment, and live in dignity for future generations [1]. In addition, from a merely economic point of view, it was also a fact that environmental problems and irresponsible activities of industries towards the environment would have caused a "boomerang effect" [2]. Therefore, LCA has emerged as an essential methodology in which the environmental impacts of a system (product or process) are identified and quantified throughout its life cycle [3]. Indeed, LCA has become a decision-making tool for industries and researchers, where outcomes support alternative seeking to issues such as substituting hazardous materials, optimizing raw materials, and identifying hotspots within processes [4]. Industrial scale LCA studies are the most common, retrospective, and change-oriented, whereas scale-up and laboratory scale studies are scarce, prospective, and future-oriented. Unfortunately, the potential of lab-scale LCA studies has been underestimated up to now since they can provide valuable inputs at the initial stages of emerging technologies, avoiding future modifications at industrial applications that would reflect high cost and time investments [5,6].

In the 1970s, a landmark achievement in photo electrocatalysis was accomplished by Fujishima and Honda, with the development of photo electrocatalytic water splitting (PEC) technology. This led to the emergence of heterogeneous photocatalysis, which has established itself as a straightforward and eco-friendly technique with high potential for energy and environmental applications. Among the semiconductors used in this field, titanium dioxide (TiO_2) has been the most important due to its low cost, non-toxicity, chemical stability, and excellent performance under UV irradiation [7]. Moreover, studies have demonstrated that other metal oxides such as ZnO, CuO, CeO_2 [8–10], and metal sulfides, including ZnS, CuS, MoS [11], as well as their heterostructures, have made significant progress in the degradation of water pollutants under sunlight, UV, and visible illumination. For instance, $ZnS/SnO_2/g\text{-}C_3N_4$ composite was investigated for methylene blue (MB) and RhB dyes degradation under visible light. This ternary hybrid showed 95 % of photocatalytic efficiency for 10 ppm RhB solution, besides 98 % degradation of 10 ppm MB solution after 25 min and 60 min, respectively. The excellent performance was attributed to the synergistic effect among ZnS quantum dots, SnO2 nanoparticles, and $g\text{–}C_3N_4$, which minimize the recombination electron and hole pairs. In addition, 32.5 wt

* Corresponding author.
E-mail address: marcela.frias@unimi.it (M.F. Ordoñez).

https://doi.org/10.1016/j.ceja.2023.100575

Available online 24 November 2023

% of CuS onto ZnS was optimum to design of an extraordinary heterojunction with the ability to thoroughly degrade 20 × 10^{-5} M MB solution within 2 min under visible light irradiation. Herein, the formation of heterojunction was effective to enhance charge carriers separation [11].

On the other hand, it is worth noting that perovskite oxides (ABO_3) are third-generation promising materials for photocatalytic processes. These materials exhibit high thermal stability and resistance to photo corrosion, making them ideal for water-splitting and solar cell applications [12,13]. Therefore, it has also been of immense scientific interest to exploit these properties for environmental remediation purposes, as pointed out by Djellabi et al. [14]. It is the case of strontium titanate ($SrTiO_3$), an attractive ceramic material to optoelectronic industry due to its high thermal stability and wide band gap (3.2 eV) analogous to TiO_2 [15-18], which number of investigations related to its use has grown steadily over the last twenty years, as it is observed in Fig. 1. Regarding $SrTiO_3$ production, multiple physical and chemical synthesis routes can be employed, for instance, solid-state reaction [15,19,20], sol–gel [21], microwave-assisted hydrothermal [22], and solvothermal [23]. Most investigations consider the evaluation of synthesis parameters and their impact on $SrTiO_3$ physical–chemical properties. Nevertheless, it is not certain that all procedures align with green chemistry principles and add sustainable value [24]. To the best of our knowledge, there is a gap in the assessment of $SrTiO_3$ production pathways and potential environmental impacts, especially if industrial manufacturing is envisaged.

In this paper, we aimed to demonstrate how laboratory-scale LCA studies can provide an early environmental profile and understanding. We accomplished this by conducting a comparative LCA analysis of six representative $SrTiO_3$ synthesis routes for the first time. The primary objective was to assess the environmental impacts associated with each synthesis process, engage in constructive discussions regarding these findings, and utilize the results to make informed decisions in selecting the most environmentally sustainable technique(s).

It is our firm belief that such an approach should be made mandatory for all companies intending to introduce new materials into production. This proactive strategy enables us to comprehend the environmental consequences at the inception of a project, rather than discovering unmanageable impacts once the production is already underway. By adopting this approach, companies can steer their endeavors towards sustainability from the very beginning, avoiding potential setbacks and costly retrofits. This not only benefits the environment but also ensures long-term economic viability and competitiveness in a world increasingly focused on sustainable practices.

2. Materials and methods

2.1. Goal and scope definition

The main goal of this study was to compare the environmental impacts of six different routes for the synthesis of $SrTiO_3$ photocatalyst. Moreover, among them, diagnose the critical steps or materials, and determine the suitable pathway(s) for eco-$SrTiO_3$ ceramic material synthesis. Synthetic procedures can be classified into two main categories based on whether or not the calcination step is required. For instance, solid-state reaction, sol–gel method, and sonochemical method are approaches where heat treatment is essential to produce a specific crystal structure. Whilst, hydrothermal, molten-salt, and solvothermal methods are bottom-up syntheses that do not involve a heat treatment as the final step. We selected a mass-based functional unit in this LCA study: 1 g of $SrTiO_3$ photocatalyst. The potential impacts were estimated for 1 g of $SrTiO_3$ produced in the laboratory. The six systems of boundaries are displayed in Fig. 2. The description of each synthesis route is explained in Section 2.2.

2.2. Description of the synthesis routes

2.2.1. Solid-state reaction method (SSR)

The SSR method is a straightforward, cost-effective, and no-waste generation technique in which the synthesis is carried out without the use of solvents [15,25]. For this reason, it is a conventional and eco-friendly mechanochemical process for several semiconducting metal oxides production at large scale [26]. Carbonates, nitrates, or hydroxides are commonly employed as precursors; extreme temperatures (1100–1300 °C) and mills (i.e., rotary or vibration ball mills) are essential at the industrial scale. However, for lab-scale purposes, the fabrication of $SrTiO_3$ was calculated based on strontium carbonate ($SrCO_3$) and titanium dioxide (TiO_2, anatase) as starting materials with a Sr/Ti ratio of 1 (Eq. (1)).

$$SrCO_3 + TiO_2 \rightarrow SrTiO_3 + CO_2 \quad (1)$$

The reagents were mixed by stirring for 15 min to simulate an agate mortar action and then calcined at 1000 °C to avoid as many impurities as possible [27], adopting the following ramp: 0.5 °C/min from 25 to 250 °C, time 1 min; 2.5 °C/min from 250 to 1000 °C, time 60 min.

2.2.2. Sol–gel route (SG)

The SG process is a wet chemical technique based on an initial colloidal suspension (sol) of metal precursors (e.g., metal alkoxides) and an integrated network (gel) formed during a condensation reaction. It is an outstanding synthesis route for the preparation of high-quality and homogeneous metal oxides at lower temperatures compared to SSR [28]. This technique involves five main steps: hydrolysis, condensation,

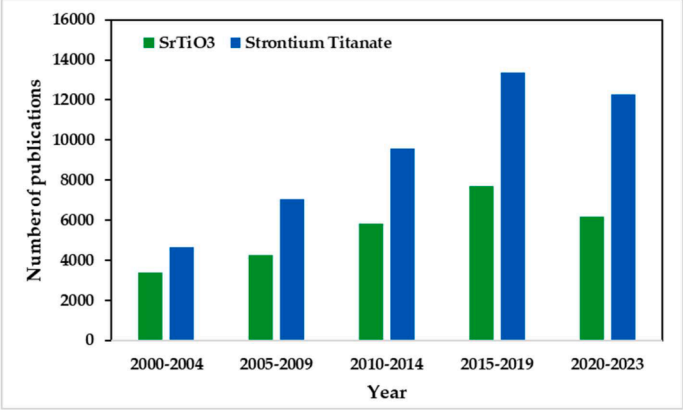

Fig. 1. Number of publications by typing keywords "$SrTiO_3$" and "strontium titanate" in the past 25 years, according to Scopus.

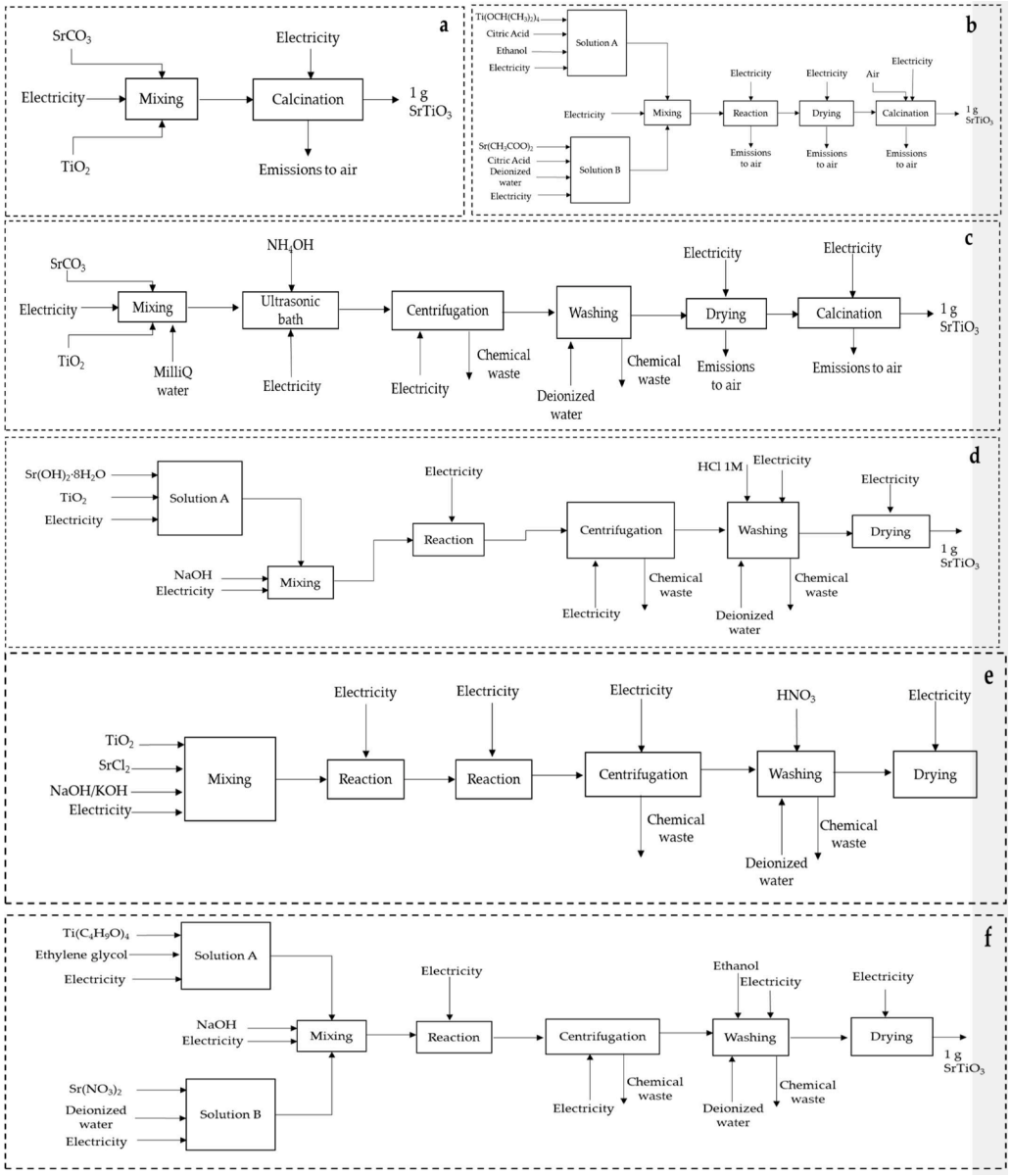

Fig. 2. System of boundaries for the six synthesis routes: (a)solid state reaction, (b)sol–gel, (c) sonochemical, (d) hydrothermal, (e) molten-salt, and (f) solvothermal method.

aging, drying, and calcination. The first two steps are strongly influenced by reaction conditions such as temperature, pH, solvent, and precursor concentration [29]. In the aging step, the formation of a polymer network continues. The time can affect the quality of your final product [30]. During the drying step, the liquid within the gel is removed. Depending on drying conditions (e.g., under ambient, supercritical, or freezing conditions) the gel is called aerogel, xerogel, or cryogel, respectively, and they exhibit different features. Lastly, in both drying and calcination, temperature plays a key role in removing surface hydroxyl groups and getting our crystalline perovskite material [31].

The SG method entails one-pot synthesis for LCA calculations, as illustrated in Fig. 2b. The entire procedure was described in detail elsewhere [32]. Briefly, titanium isopropoxide (TTIP) was added into ethanol, followed by citric acid in a molar ratio of 2:1 concerning Ti-ions to get solution A. Separately, solution B was composed of strontium acetate in distilled water and citric acid dissolved in a 2:1 molar ratio concerning Sr-ions. Then, both solutions were mixed for 3 h under vigorous at room temperature. Afterward, the solution was heated to 100 °C for 2 h to form the gel. In this step, alcohol and partial water were evaporated. The obtained gel was dried overnight at 60 °C for 15 h, followed by 9 h at 120 °C. Lastly, calcination at 1000 °C was carried out to achieve the final photocatalyst crystalline structure using the previously mentioned ramp. The main chemical reactions that took place in the synthesis of $SrTiO_3$ by the SG method are described below [33]:

$$SrC_4H_6O_4 + 2C_6H_8O_7 \rightarrow Sr(C_6H_6O_7) + 2\,CH_3COOH + C_6H_8O_7 \quad (2)$$

$$TiC_{12}H_{28}O_4 + 2C_6H_8O_7 \rightarrow Ti(C_6H_6O_7)(C_6H_{14}O_2) + 2\,C_3H_8O + C_6H_8O_7 \quad (3)$$

$$Ti(C_6H_6O_7)(C_6H_{14}O_2) + 13.5\,O_2 \rightarrow TiO_2 + 12CO_2 + 10H_2O \quad (4)$$

$$Sr(C_6H_6O_7) + 4.5\,O_2 \rightarrow SrO + 6CO_2 + 3H_2O \quad (5)$$

$$C_6H_8O_7 + 4.5\,O_2 \rightarrow 6CO_2 + 4H_2O \quad (6)$$

$$SrO + TiO_2 \rightarrow SrTiO_3 \quad (7)$$

2.2.3. Sonochemical method (US)

The US method, as well as the SSR method, is known as a cost-effective and green technique for the synthesis of perovskite oxide–based materials [34]. The employment of ultrasonic radiation leads to the generation, growth, and implosive collapse of bubbles inducing high-pressure conditions suitable for carrying out chemical reactions and improving mass transfer, reaction time, and reaction temperature for manufacturing new materials [30,31].

To prepare the $SrTiO_3$ photocatalyst by ultrasound-assisted synthesis, $SrCO_3$, and TiO_2 were selected as precursors. Both reagents were added in stoichiometric amounts in MilliQ water to form a mixture according to Eq. (1). The previous was stirred at room temperature for 15 min, followed by NH_4OH addition to reach a pH of 10. Then, the ultrasound probe was introduced and set up to 700 W with vigorous magnetic stirring for 3 h. Afterward, the product is centrifuged, washed with distilled water 4 times, and dried overnight for 12 h. Lastly, the powder must be calcined at 1000 °C based on early experiments to get the final $SrTiO_3$ structure.

2.2.4. Hydrothermal method (HT)

Hydrothermal synthesis is a commonly bottom-up technique for the preparation of ceramic compounds. This method's core relies on reactions in aqueous medium through multiple phases and at high temperatures and pressures. Therefore, to reach these conditions, a closed reaction vessel (autoclave) is essential [35]. Examining parameters like reaction temperature and time, pH, and concentration of reactants must be considered to control final particle size, chemical composition, and morphology [11,30,31].

For this work, we were based on $SrTiO_3$ hydrothermal synthesis reported elsewhere [36]. Herein, TiO_2 and $Sr(OH)_2{\cdot}8H_2O$ were mixed in an equal molar ratio. Meanwhile, 50 mL of 50 M NaOH solution was added to the mixture. The final mix was transferred to a Teflon-lined stainless-steel autoclave and heated at 220 °C for 20 h. After the reaction, the mixture was centrifuged and washed with deionized water several times and HCL 1 M to eliminate NaOH excess and the presence of carbonates, respectively. Then, the slurry was passed again with deionized water until it reached a neutral pH. The final precipitate was dried overnight at 80 °C. The main chemical reaction is displayed in Eq. (8).

$$Sr(OH)_2{\cdot}8H_2O + TiO_2 \rightarrow SrTiO_3 + 9H_2O \quad (8)$$

2.2.5. Solvothermal method (SV)

The SV method is similar to the HT method. Chemical reactions occur within an autoclave at high pressure and temperatures in non-aqueous solutions (e.g., use of ethanol or polyols). Since photocatalytic properties of our final perovskite can be enhanced by modifying particle size, shape, and morphology, this method offers the production of high-quality materials with a narrow size distribution, besides vasty morphologies, and structures that with conventional methods at high temperatures are not possibly reached [36,37]. Many studies have reported using solvothermal synthesis to prepare perovskite oxides with applications such as photocatalysis and energy storage/conversion [38,39]. Nevertheless, the number of steps or the use of organic solvents are some of the issues that industries deal with to scale up the process since it increases expenses and greenhouse gases [40].

For this LCA study, $SrTiO_3$ synthesis via the SV method reported by Li et al. [41] was employed. Briefly, tetra-n-butyl titanate (TBT) was added dropwise into ethylene glycol under magnetic stirring at ambient conditions to form solution A. Separately, solution B was composed of strontium nitrate, and NaOH was added to distilled water. Then, solution A was added into solution B bearing in mind a vigorous stirring for 15 min. Afterward, the obtained mixture was transferred into a Teflon-lined stainless-steel autoclave and heated at 150 °C for18 h. The slurry was centrifuged, washed several times with distilled water and ethanol, and dried overnight for 12 h to obtain pure $SrTiO_3$. The chemical reactions considered in the synthesis are described in Eq (9)–12.

$$Ti(OCH_2CH_2CH_2CH_3)_4 + C_2H_6O_2 \rightarrow Ti(OCH_2CH_2CH_2CH_3)_4 + C_2H_6O_2 \quad (9)$$

$$Sr(NO_3)_2 + 2NaOH \rightarrow Sr(OH)_2 + 2NaNO_3 \quad (10)$$

$$Ti(OCH_2CH_2CH_2CH_3)_4 + 2H_2O \rightarrow TiO_2 + 4\,CH_3CH_2CH_2CH_2OH \quad (11)$$

$$Sr(OH)_2 + TiO_2 \rightarrow SrTiO_3 + H_2O \quad (12)$$

2.2.6. Molten salt method (MS)

Molten salt is reported as a simple, cost-effective, and easily scale-up technique for synthesizing nanomaterials. In this process, metal precursors are mixed with a eutectic mixture (e.g., NaOH/KOH), followed by a heating process up to the melting point of salts, and the formation of the final ceramic material starts through precipitation. Additional salts are removed during the washing step with water [42].

The molten salts act as solvents, favoring the reaction rate, and increasing the contact area between reagents, besides reducing the working temperature, controlling the size and morphology, and avoiding agglomeration. Therefore, as well as HT method parameters such as reaction temperature, annealing time, pH, type of precursors, and concentration play a key role in the synthesis process [40].

The synthesis of $SrTiO_3$ reported by Liu et al. [43] was used for LCA calculations. Briefly, a mixture of hydroxides (NaOH/KOH 51.5:48.5) was placed in a Teflon vessel. Then, $SrCl_2$ and TiO_2 were mixed, added onto the vessel, and heated to 205 °C for 3 h to thoroughly melt hydroxides. After 4 days of reaction, the vessel was cooled down, and the mixture was centrifuged. Subsequently, it was washed several times with distilled water and diluted nitric acid to avoid hydroxides excess. Lastly, the white powder was dried overnight to obtain our final product. The synthesis of $SrTiO_3$ through the molten-salt route is described in Eq. (13)–19, Eq. (19) the overall reaction:

$$2NaOH + TiO_2 \rightarrow Na_2TiO_3 + H_2O \quad (13)$$

$$2KOH + TiO_2 \rightarrow K_2TiO_3 + H_2O \quad (14)$$

$$SrCl_2 + 2NaOH \rightarrow Sr(OH)_2 + 2NaCl \quad (15)$$

$$SrCl_2 + 2KOH \rightarrow Sr(OH)_2 + 2KCl \quad (16)$$

$$Sr(OH)_2 + Na_2TiO_3 \rightarrow SrTiO_3 + 2NaOH \quad (17)$$

$$Sr(OH)_2 + K_2TiO_3 \rightarrow SrTiO_3 + 2KOH \quad (18)$$

$$SrCl_2 + TiO_2 + NaOH + KOH \rightarrow SrTiO_3 + NaCl + KCl + H_2O \quad (19)$$

2.3. Life cycle inventory and LCA modeling

Life cycle assessment analysis was performed using SimaPro 9.4 software. Data related to the syntheses was provided from laboratory experiments and literature, whereas background data was available in the Ecoinvent 3.8 database Table 1). Sub-processes belonging to the production of some reagents were created since data was nonexistent. Based on literature and patents the final reactions were considered (Eqs. 20–(25). Concerning electricity production, all synthesis routes included medium voltage electricity production from Italy in 2014.

This cradle-to-gate assessment considered resource extraction, material processing, and production of 1 g of $SrTiO_3$ photocatalyst (function unit); therefore, the by-products were handled as emissions to air or waste to be chemically treated. The use and end-of-life phases should have been addressed. A detailed life cycle inventory from synthesis routes modeling is described in Table S1. Lastly, the impact categories were estimated conforming to the CML baseline method (Table 2),

Table 1
Inventory data employed from the Ecoinvent database.

Acetic acid	Acetic acid, without water, in 98 % solution state {GLO}/ market for
NH_3	Ammonia, anhydrous, liquid {RER} / market for ammonia, anhydrous, liquid
Citric acid	Citric acid {GLO} / market for
Electricity	Electricity, medium voltage {IT} / market for
Ethylene glycol	Ethylene glycol {GLO} / market for
Ethanol	Ethanol, without water, in 99.7 % solution state, from ethylene {RER} / market for ethanol, without water, in 99.7 %
HCl	Hydrochloric acid, without water, in 30 % solution state {RER} / market for
Isopropanol	Isopropanol {RER} / market for isopropanol
Nitric acid	Nitric acid, without water, in 50 % solution state {RoW} / market for nitric acid, without water, in 50 % solution state
KOH	Potassium hydroxide {GLO} / market for
NaOH	Sodium hydroxide, without water, in 50 % solution state {RoW} / market for
$SrCO_3$	Strontium carbonate {GLO} / market for
TiO_2	Titanium dioxide {GLO} / market for
$TiCl_4$	Titanium tetrachloride {GLO} / market for
Distilled water	Water, deionized {Europe without Switzerland} / market for water, deionized
MilliQ water	Water, ultrapure {RER} / market for water, ultrapure

Table 2
Midpoint impact categories used for the evaluation method (CML baseline).

Impact category group	Impact category name	Unit
Depletion of abiotic resources	Abiotic depletion (elem., ultimate reserves)	kg Sb eq
	Abiotic depletion (elem., fossil fuels)	kg Sb eq
Climate change	Global warming 100a (incl. NMVOC av.)	kg CO_2 eq
Ozone layer depletion	Ozone layer depletion - ODP steady state	kg CFC-11 eq
Ecotoxicity	Freshwater aquatic ecotoxicity - FAETP inf	kg 1,4-DB eq
	Marine aquatic ecotoxicity - MAETP inf	kg 1,4-DB eq
	Terrestrial ecotoxicity - TETP inf	kg 1,4-DB eq
Human Toxicity	Human toxicity - HTP inf	kg 1,4-DB eq
Photochemical oxidation	Photochemical oxidation (high NOx)	kg C2H4 eq
Acidification	Acidification potential - average Europe	kg SO2 eq
Eutrophication	Eutrophication - generic	kg PO4—eq

corresponding to the most common impact categories used in LCA [44]. Carbon and Water footprint were also assessed as a single issue using ICCP (kg CO_2-eq) and AWARE method (m^3) method, respectively.

$$TiCl_4 + 4C_4H_{10}O \rightarrow TiC_{16}H_{36}O_4 + 4HCl \quad (20)$$

$$TiCl_4 + 4C_3H_8O \rightarrow Ti\left(OCH(CH_3)_2\right)_4 + 4HCl \quad (21)$$

$$SrCO_3 + 2HNO_3 \rightarrow Sr(NO_3)_2 + H_2O + CO_2 \quad (22)$$

$$2CH_3COOH \rightarrow Sr(CH_3COO)_2 + H_2O + CO_2 \quad (23)$$

$$SrCO_3 + 2HCl \rightarrow SrCl_2 + H_2O + CO_2 \quad (24)$$

$$SrCl_2 + 2NaOH + 8H_2O \rightarrow Sr(OH)_2{\cdot}8H_2O + 2NaCl \quad (25)$$

3. Results and discussion

The results and discussion of this work have been organized into four sections: Section 3.1 discusses a comparative LCA study of titanium (Ti) and strontium (Sr) precursors. Then, a comparative LCA study of the six synthesis routes and the identification of key steps were explained in Section 3.2. Lastly, carbon footprint and water scarcity footprint were discussed in Sections 3.3 and 3.4, respectively.

3.1. Comparative environmental analysis of reagents

As a starting point, a comparative analysis among strontium and titanium precursors was performed to comprehend the initial environmental burden of raw materials. The results of characterization are shown in Fig. 3 and Table S2. As can be seen, among titanium precursors, titanium tetrachloride ($TiCl_4$) exhibits a significant contribution in different categories such as global warming, ozone layer depletion, human toxicity, freshwater and marine aquatic ecotoxicity, and terrestrial ecotoxicity. The burden could be associated with the chlorination and refining process required in the production of $TiCl_4$ [45]. On the other hand, among strontium precursors, strontium hydroxide octahydrate ($Sr(OH)_2{\cdot}8H_2O$) causes significant negative effects in all categories except for photochemical oxidation. Significant environmental impacts are related to strontium carbonate ($SrCO_3$) production, which is the first rung for $Sr(OH)_2{\cdot}8H_2O$ synthesis, and demands $SrSO_4$ as raw material in the black ash process [46]. Therefore, all upstream processes and synthesis routes that use TBT, TTIP, and $Sr(OH)_2{\cdot}8H_2O$ would drag with the load.

Furthermore, the normalization results are displayed in Fig. 4 and Table S3. Normalization results enable us to compare the impact categories and understand their relevance. Thus, it is observed that marine aquatic ecotoxicity is the most impacted environmental category, in the order $TiCl_4$, > TTIP > TBT > TiO_2. The titanium chloride production process, a raw material for TTIP and TBT synthesis, contributes to essential oxygen furnace slag generation, a by-product of the steelmaking industry. In contrast, among strontium precursors: $Sr(OH)_2{\cdot}8H_2O$ substance is a significant contributor, followed by strontium chloride ($SrCl_2$), strontium acetate ($Sr(C_2H_3O_2)_2$), strontium nitrate ($Sr(NO_3)_2$), and $SrCO_3$. Herein, the environmental burdens are dominated by $SrCO_3$ and HCl production processes.

3.2. Comparative environmental analysis of synthesis routes

The synthesis routes with and without heat treatment (i.e., calcination step) were assessed separately to identify their hotspots and the most affected categories and determine which processes were ecological or contributed the most to the environmental pollution of each group. The characterization and normalization results of calcination methods for synthesizing $SrTiO_3$ photocatalyst (SSR, US, SG) are shown in Fig. 5. From the characterization results (Fig. 5 (left) and Table S4), undoubtedly, SG is the major contributor to all environmental impact categories (67–74 %), followed by in equal proportion (16 %) US and SSR. Nevertheless, according to normalization results (Fig. 5 (right) and Table S5), marine aquatic ecotoxicity potential exhibited the highest score. Its environmental burden was associated with electricity production procedures.

Therefore, it was essential to determine the SG hotspot, the stage dominating most environmental load. A comparison analysis of all the phases involved during the synthesis was performed (Fig. 6 and Table S6). The calcination step dominated environmental impacts. Therefore, decisions should be made at this stage to reduce environmental effects, mainly marine aquatic ecotoxicity.

On the other hand, Fig. 7 (left) and Table S7 exhibit the characterization results for non-calcination synthesis routes (HT, MS, SV). All the environmental impact categories were equally affected by MS and HT. From normalization results (Fig. 7(right) and Table S8), it was evident that MS and HT had by far the most significant environmental impact contribution in marine aquatic ecotoxicity (46 % and 36 %, respectively). Moreover, freshwater aquatic ecotoxicity potential, abiotic depletion potential (from fossil fuels), global warming potential,

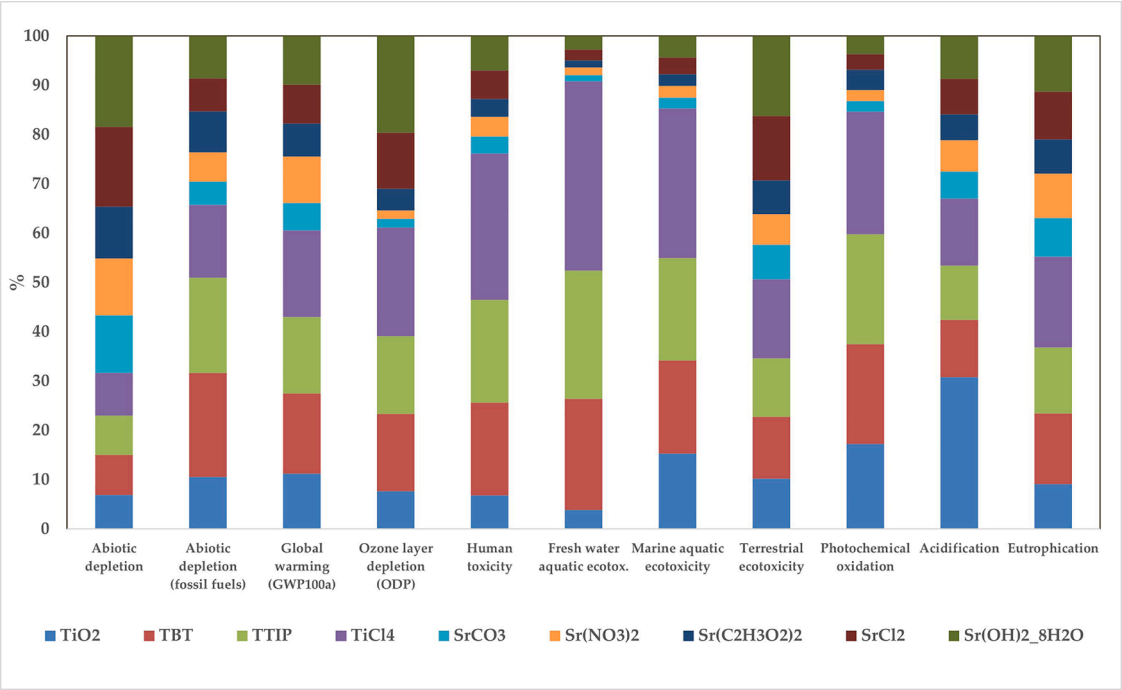

Fig. 3. Characterization results of Sr and Ti precursors environmental cycle assessment.

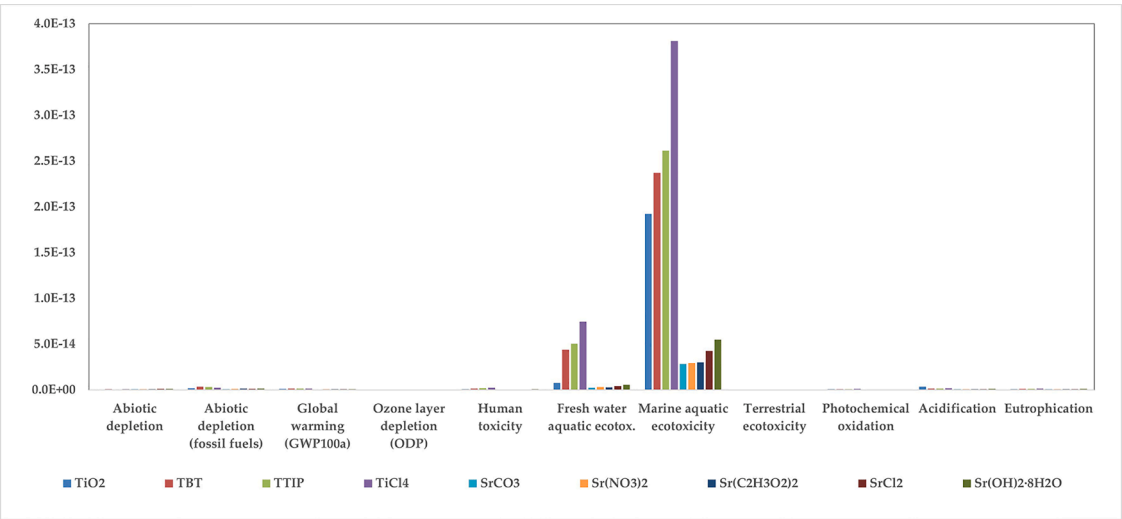

Fig. 4. Normalization results of Sr and Ti precursors environmental cycle assessment.

acidification, and eutrophication potential were lower than it. Electricity requirement was identified as a significant contributor to these categories since the burden was linked to electricity production. Lastly, SV could be considered an environmentally friendly synthesis route among non-calcination approaches.

Herein, finding out the step with high energy consumption within MS was also crucial. The results in Fig. 8 and Table S9 demonstrated that washing and centrifugation were energy-demanding stages. Nevertheless, the drying step required 10 % more energy, being the key step. In this matter, drying time is an essential parameter that directly affects the environmental response. To solve this issue, some studies determined that the drying times get shorter by increasing the drying air temperature [47]. Thus, a balance between the time and drying air temperature should be reached that would not compromise the structure of our final $SrTiO_3$ photocatalyst.

To close this sub-LCA study, we compared the most eco-friendly approaches of each group: US and SSR from calcination approaches and SV from non-calcination. The characterization and normalization results for this comparative study are displayed in Fig. 9 and Table S10 and 11.

The obtained LCA characterization results revealed that SV was the largest contributor in all environmental impact categories due to electricity consumption during drying (79 %), without discarding its requirement during the washing and centrifugation stages. Based on normalization results, marine aquatic ecotoxicity was the dominant environmental impact category among the three synthesis processes. Nevertheless, other impact categories such as freshwater aquatic ecotoxicity, abiotic depletion (from fossil fuels), global warming, acidification, and eutrophication were also significant. The normalization results also demonstrated that SSR and US methods have the lowest environmental effects. The use of eco-raw materials such as $SrCO_3$ and TiO_2 could have contributed to reducing the initial environmental inputs

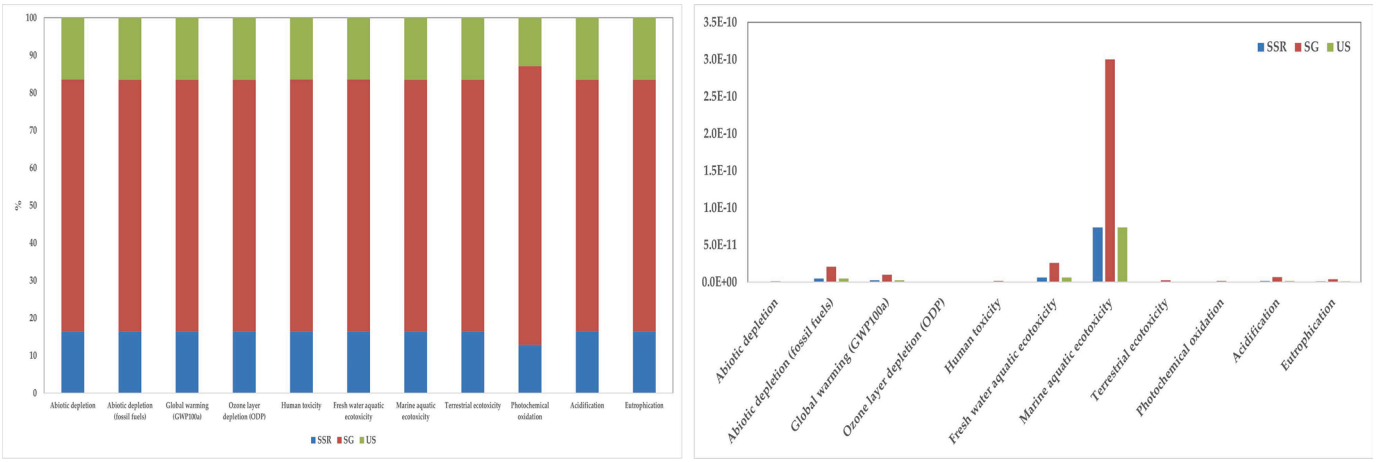

Fig. 5. Characterization results (left) and normalization results (right) of synthesis methods with calcination step (SSR, SG, US).

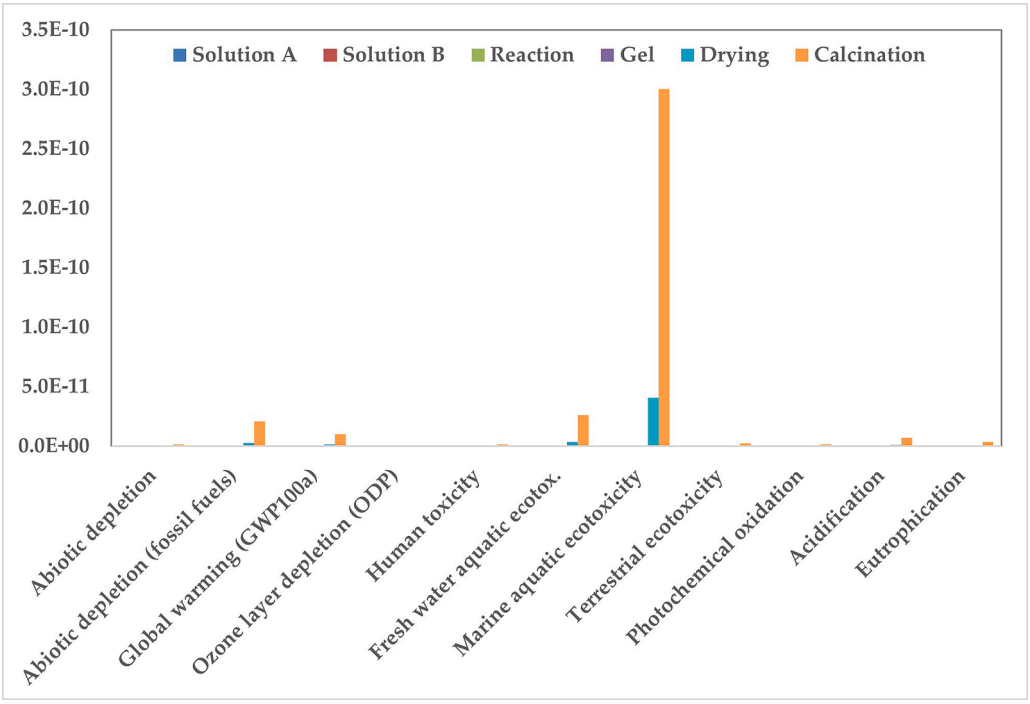

Fig. 6. Normalization results SG method critical step assessment.

since the energy consumption factor was revealed as the driving force of significant environmental impacts. Although the difference is minimal (0.37 %), the US requires more electricity than SSR. Based on the previous results, since the most contaminating method for $SrTiO_3$ production was a non-calcination approach (i.e. SV), it is worth noting that the calcination step was less polluting than the drying stage. Lastly, this work was performed using Italian electricity, so future studies could be addressed to evaluate these synthesis procedures in other countries and use cleaner resources for electricity that might effectively reduce life cycle impacts.

3.3. Global warming potential assessment

In the previous comparative LCA study, marine aquatic ecotoxicity was the most significant environmental impact. Therefore, it was crucial to analyze separately global warming potential for the next 20 years through the IPCC method (Intergovernmental Panel on Climate Change). The global warming indicator calculates the contribution of gaseous emissions released during SrTiO3 synthesis processes to climate change issues [48].

Fig. 10 (left) and Table S12 show characterization results. Interestingly, it can be observed that the main contributors to global warming were all three synthesis routes: MS, HT, and SV, in descending order. In addition, without discriminating the source of CO_2 molecules (i.e., fossil fuels, biogenic, or land transformation), the behavior among non-calcination methods was similar: MS process contributed 6 % more to CO_2 emissions than HT, and 29 % more than SV. On the other hand, among calcination synthesis routes, SG contributes approximately 7 % to GWP, followed by SSR (2 %) and US (2 %). Therefore, from the GWP point of view and to produce eco-$SrTiO_3$ materials with the least amount of pollutant gas emissions to the atmosphere, approaches involving a final calcination stage such as solid-state, ultrasound-assisted, and sol–gel reaction should be employed instead of non-calcination routes.

Eventually, considering the damage assessment (Fig. 10(right), Table S13), most of the global warming contribution in MS synthesis came from CO_2 and CH_4 molecules originating from fossil fuels or

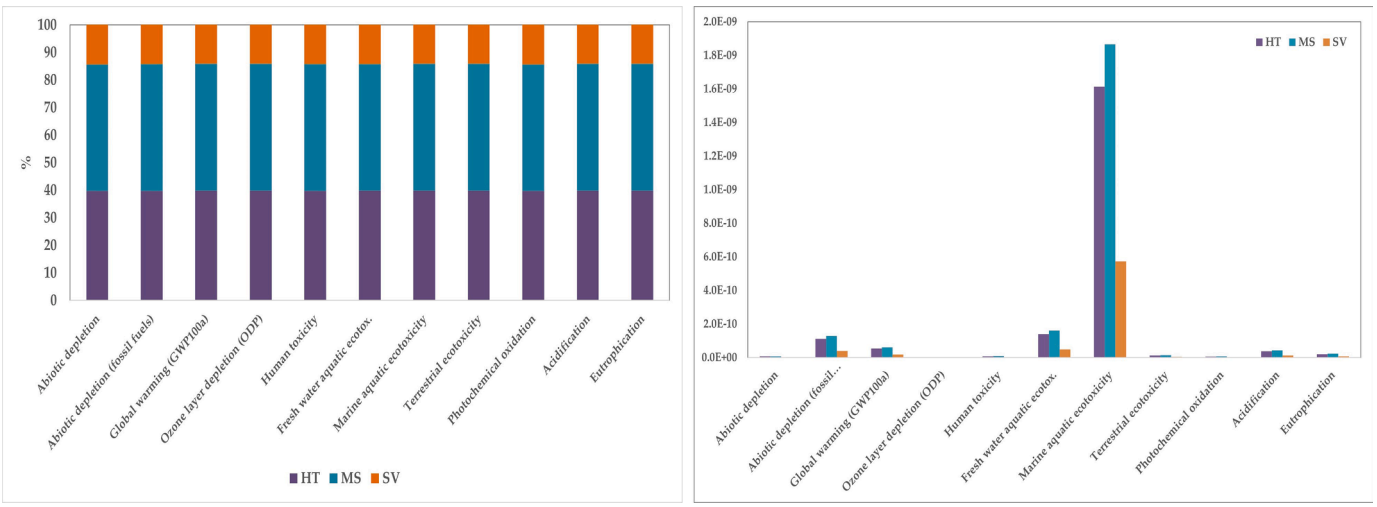

Fig. 7. Characterization results (left) and normalization results (right) of synthesis methods without calcination step (HT, MS, SV).

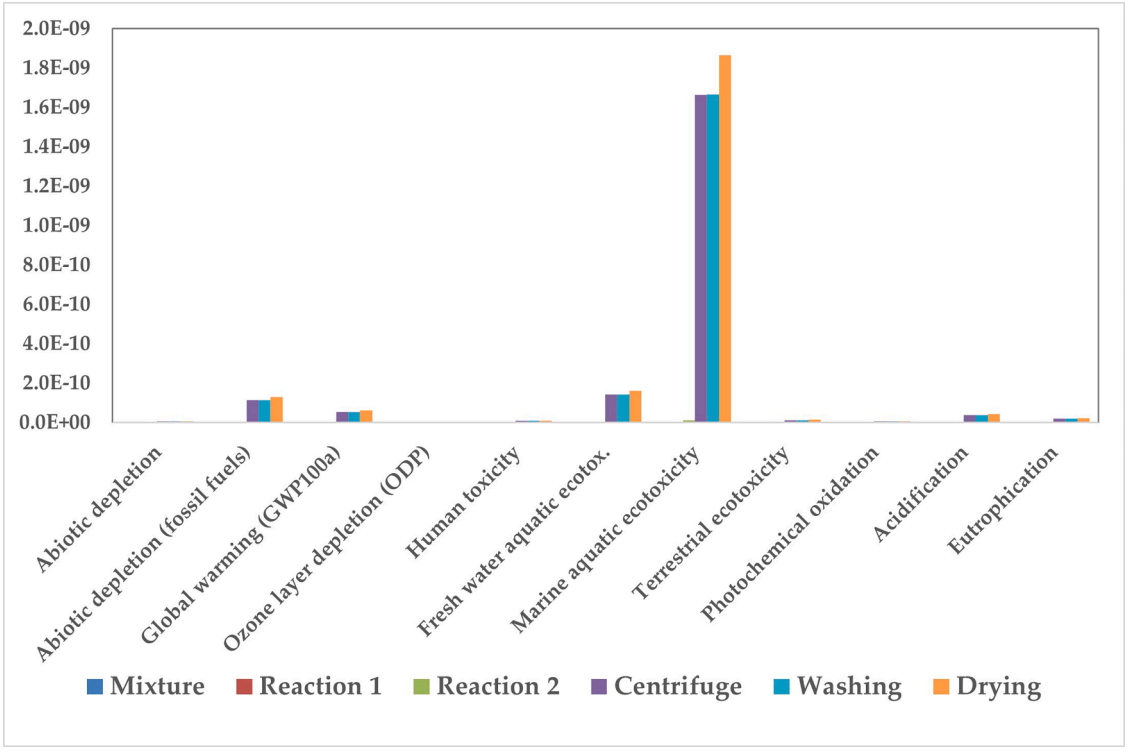

Fig. 8. Normalization results MS method critical step assessment.

biogenic sources. The former was mainly linked to the electricity production process in an average hard coal power plant or a combined cycle power plant with natural gas. At the same time, the latter was equivalented to CH_4 losses from biogas production plants.

Finally, it is worth noting that this work considered Italian medium voltage electricity as energy input. Therefore, further investigations are encouraged to assess different electricity sources and site locations to analyze GWP and other environmental impact categories influenced by energy factors.

3.4. Water footprint

The AWARE calculation methodologies were used to assess the water scarcity footprint (i.e., "the available water remaining" [49]. Fig. 11 and Table S14 illustrate the contribution of each synthesis process to water scarcity. MS and HT showed the greatest water scarcity impacts influenced by the amount of water employed in the electricity production process at grid-connected reservoir hydropower plants. On the other hand, among calcination synthesis routes, SG showed the greatest water scarcity footprint. Nevertheless, it is worth noting that MS's impact was six times higher than SG's. Moreover, since water scarcity footprint was also linked to energy consumption factor, non-calcination processes would not be a good decision for the synthesis of eco-$SrTiO_3$ materials. In contrast, SSR and US were considered ecological, with less than 5 % of the environmental burden contribution.

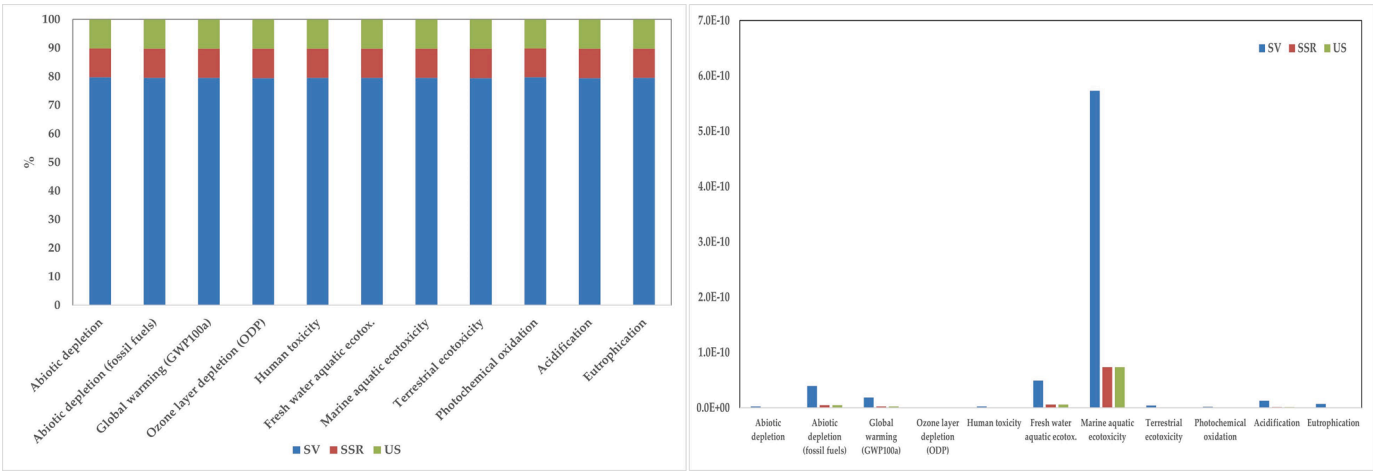

Fig. 9. Characterization results (left) and normalization results (right) of comparison eco-friendly synthesis routes (SV, SSR, US).

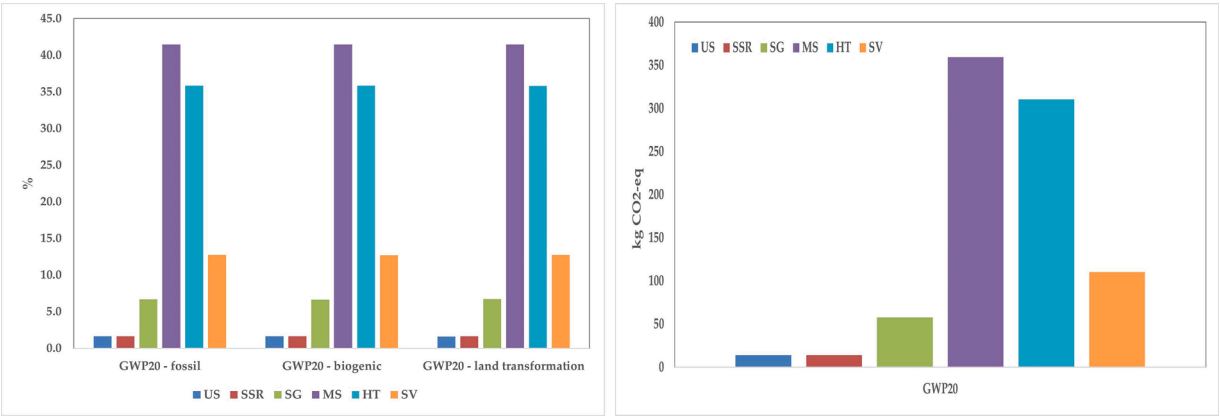

Fig. 10. Characterization results (left) and damage assessment results (right) for global warming potential analysis.

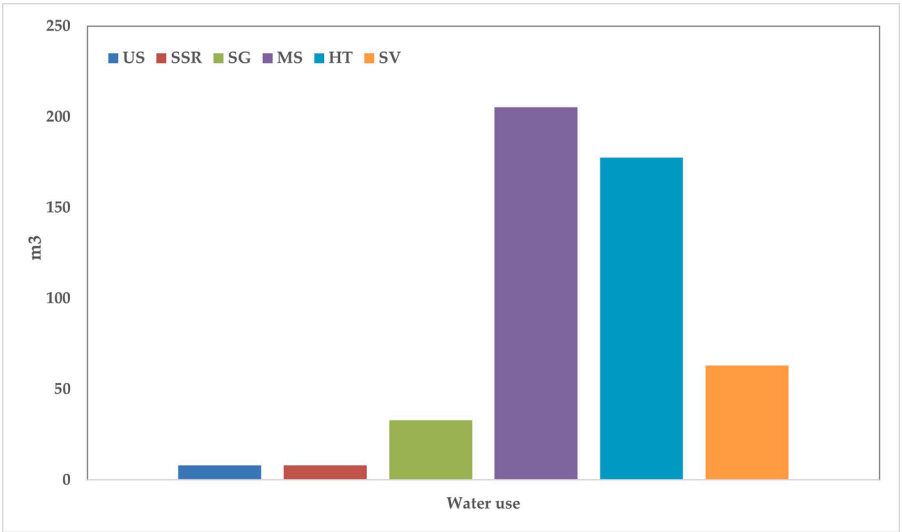

Fig. 11. Results of water scarcity footprint assessment.

4. Conclusions

$SrTiO_3$ is a promising ceramic material with applications in various fields, including photocatalysis. However, it is essential to recognize the urgent need for responsible environmental management in today's world, where the sustainability of our economic activities and their resulting products is paramount. To ensure the eco-friendly industrial production of $SrTiO_3$ on a large scale, it is crucial to proactively identify and mitigate potential environmental and health impacts associated with different manufacturing processes.

In this groundbreaking LCA study, we have established a baseline understanding of the environmental effects of six laboratory-scale $SrTiO_3$ synthesis routes. Among the various titanium and strontium precursors considered, TiO_2 and $SrCO_3$ emerged as the least environmentally impactful options. Conversely, some reagents, notably $TiCl_4$ and $SrCl_2$, were found to have significant environmental burdens associated with their upstream lifecycle production stages.

The LCA environmental assessment revealed that the MS synthesis route exhibited the highest environmental impacts among non-calcination processes, primarily due to electricity consumption, with the drying step identified as a hotspot. Similarly, the SG route proved to be the most environmentally impactful, with the calcination stage being the dominant contributor to environmental impacts. Furthermore, our analysis of global warming potential (GWP) and water scarcity footprint indicated that SSR and US were the most sustainable and suitable production processes for eco-friendly $SrTiO_3$ materials.

Future research efforts should extend to a cradle-to-grave LCA study, encompassing both laboratory-scale and scale-up production, with a focus on the use and end-of-life stages. Additionally, the exploration of renewable energy sources is critical, as a substantial portion of CO_2 emissions was linked to fossil energy use. This holistic approach will be essential in achieving the goal of producing $SrTiO_3$ materials that are not only technologically advanced but also environmentally responsible.

Funding statement

The authors received no financial support for this research, authorship, and/or publication of this article.

CRediT authorship contribution statement

Marcela Frias Ordoñez: Data curation, Formal analysis, Writing – review & editing, Supervision. **Serena Biella:** Data curation, Formal analysis, Investigation, Methodology, Writing – original draft. **Ermelinda Falletta:** Conceptualization, Data curation, Funding acquisition, Project administration, Supervision, Writing – review & editing. **Claudia L. Bianchi:** Data curation, Formal analysis, 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.

Data availability

Data will be made available on request.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ceja.2023.100575.

References

[1] B. N. Singh and B. R. K. Sinha, Quality of Life of Slum Dwellers: A Theoretical Approach. 2019.
[2] M. Petrović-Ranđelović, T. Stevanović, M. Ivanović-Đukić, Impact of Corporate Social Responsibility on the Competitiveness of Multinational Corporations, Procedia Econ. Financ. 19 (15) (2015) 332–341, https://doi.org/10.1016/s2212-5671(15)00034-9.
[3] B. Godskesen, N. Meron, and M. Rygaard, LCA of drinking water supply. 2017.
[4] J. Pryshlakivsky, C. Searcy, Life Cycle Assessment as a decision-making tool: Practitioner and managerial considerations, J. Clean. Prod. 309 (2021), 127344, https://doi.org/10.1016/j.jclepro.2021.127344. February 2020.
[5] M. Buyle, A. Audenaert, P. Billen, K. Boonen, S. Van Passel, The future of ex-ante LCA? Lessons learned and practical recommendations, Sustain. 11 (19) (2019) 1–24, https://doi.org/10.3390/su11195456.
[6] R. Arvidsson, et al., Environmental Assessment of Emerging Technologies: Recommendations for Prospective LCA, J. Ind. Ecol. 22 (6) (2018) 1286–1294, https://doi.org/10.1111/jiec.12690.
[7] K. Ahmad, H.R. Ghatak, S.M. Ahuja, A review on photocatalytic remediation of environmental pollutants and H2 production through water splitting: A sustainable approach, Environ. Technol. Innov. 19 (2020), 100893, https://doi.org/10.1016/j.eti.2020.100893.
[8] S.R. Mishra, M. Ahmaruzzaman, CuO and CuO-based nanocomposites: Synthesis and applications in environment and energy, Sustain. Mater. Technol. 33 (2022) e00463, https://doi.org/10.1016/j.susmat.2022.e00463. May.
[9] M. Ahmaruzzaman, Metal oxides (ZnO, CuO and NiO)-based nanostructured materials for photocatalytic remediation of organic contaminants, Nanotechnol. Environ. Eng. 8 (1) (2023) 219–235, https://doi.org/10.1007/s41204-022-00284-8.
[10] E. Kusmierek, A CeO2 semiconductor as a photocatalytic and photoelectrocatalytic material for the remediation of pollutants in industrial wastewater: A review, Catalysts 10 (12) (2020) 1–54, https://doi.org/10.3390/catal10121435.
[11] V. Gadore, S.R. Mishra, M. Ahmaruzzaman, Metal sulphides and their heterojunctions for photocatalytic degradation of organic dyes-A comprehensive review, 30, Springer, Berlin Heidelberg, 2023.
[12] M. Neophytou, et al., Enhancing the Charge Extraction and Stability of Perovskite Solar Cells Using Strontium Titanate (SrTiO3) Electron Transport Layer, ACS Appl. Energy Mater. (2019), https://doi.org/10.1021/acsaem.9b01567.
[13] A. Kumar, A. Kumar, V. Krishnan, Perovskite Oxide Based Materials for Energy and Environment-Oriented Photocatalysis, ACS Catal 10 (17) (2020) 10253–10315, https://doi.org/10.1021/acscatal.0c02947.
[14] R. Djellabi, M.F. Ordonez, F. Conte, E. Falletta, C.L. Bianchi, I. Rossetti, A review of advances in multifunctional XTiO3 perovskite-type oxides as piezo-photocatalysts for environmental remediation and energy production, J. Hazard. Mater. 421 (2022), 126792, https://doi.org/10.1016/j.jhazmat.2021.126792. May 2021.
[15] N. Sharma, K. Hernadi, The Emerging Career of Strontium Titanates in Photocatalytic Applications: A Review, Catalysts 12 (12) (2022) 1–16, https://doi.org/10.3390/catal12121619.
[16] Y. Liu, et al., Synthesis and high photocatalytic hydrogen production of SrTiO3 nanoparticles from water splitting under UV irradiation, J. Power Sources 183 (2) (Sep. 2008) 701–707, https://doi.org/10.1016/j.jpowsour.2008.05.057.
[17] M. Morgenbesser, et al., SrTiO3 based high temperature solid oxide solar cells: Photovoltages, photocurrents and mechanistic insight, Solid State Ionics 368 (2021), 115700, https://doi.org/10.1016/j.ssi.2021.115700. July.
[18] L.F. Da Silva, et al., An Understanding of the Photocatalytic Properties and Pollutant Degradation Mechanism of SrTiO3 Nanoparticles, Photochem. Photobiol. 92 (3) (2016) 371–378, https://doi.org/10.1111/php.12586.
[19] E.Rosa Silva, M. Curi, J.G. Furtado, H.C. Ferraz, A.R. Secchi, The effect of calcination atmosphere on structural properties of Y-doped SrTiO 3 perovskite anode for SOFC prepared by solid-state reaction, Ceram. Int. 45 (8) (2019) 9761–9770, https://doi.org/10.1016/j.ceramint.2019.02.011.
[20] P. Gwóźdź, A. Łącz, E. Drożdż, Effect of calcium addition on microstructure, structure, and electrical properties of SrTiO3-based materials synthesized by citrate and solid-state reaction method, Ceram. Int. 49 (10) (2023) 16332–16340, https://doi.org/10.1016/j.ceramint.2023.01.235.
[21] T. Hussain, M. Junaid, H.A. Qayyum, Preparation of Ba-doped SrTiO3 photocatalyst by sol-gel method for hydrogen generation, Chem. Phys. Lett. 754 (2020), 137741, https://doi.org/10.1016/j.cplett.2020.137741. April.
[22] R. López-Juárez, T.J. Pérez-Juache, O. Hernández-Cristobal, N. Razo-Pérez, S. Cipagauta, Microwave-hydrothermal synthesis and photocatalytic properties of Cr-doped SrTiO3 powders, Ceram. - Silikaty 65 (2) (2021) 113–119, https://doi.org/10.13168/cs.2021.0009.
[23] S. Huh, et al., Solvothermal Synthesis and Interfacial Magnetic Interaction of β-FeSe/SrTiO3-x Nanocomposites, ChemistrySelect 5 (30) (2020) 9517–9522, https://doi.org/10.1002/slct.202000871.
[24] R.A. Sheldon, Metrics of Green Chemistry and Sustainability: Past, Present, and Future, ACS Sustain. Chem. Eng. 6 (1) (2018) 32–48, https://doi.org/10.1021/acssuschemeng.7b03505.
[25] M. Jug, P.A. Mura, Grinding as solvent-free green chemistry approach for cyclodextrin inclusion complex preparation in the solid state, Pharmaceutics 10 (4) (2018), https://doi.org/10.3390/pharmaceutics10040189.
[26] S.A. Patil, et al., A simple, room temperature, solid-state synthesis route for metal oxide nanostructures, J. Mater. Chem. A 2 (33) (2014) 13519–13526, https://doi.org/10.1039/c4ta02267j.
[27] L. Chun Yan, J. Hassan, M. Hashim, W. Swee Yin, T. Foo Khoon, W.Yick Jeng, Effect of Sintering Temperatures on the Microstructure and Dielectric Properties of SrTiO 3_재백, World Appl. Sci. J. 15 (11) (2011) 1614–1618.
[28] D. Bokov, et al., Nanomaterial by Sol-Gel Method: Synthesis and Application, Adv. Mater. Sci. Eng. 2021 (2021), https://doi.org/10.1155/2021/5102014.
[29] M. Parashar, V.K. Shukla, R. Singh, Metal oxides nanoparticles via sol–gel method: a review on synthesis, characterization and applications, J. Mater. Sci. Mater. Electron. 31 (5) (2020) 3729–3749, https://doi.org/10.1007/s10854-020-02994-8.
[30] J. Li, D. Yang, X. Zhu, Effects of aging time and annealing temperature on structural and optical properties of sol-gel ZnO thin films, Aip Adv 7 (6) (2017).
[31] D. Navas, S. Fuentes, A. Castro-Alvarez, E. Chavez-Angel, Review on Sol-Gel Synthesis of Perovskite and Oxide Nanomaterials, Gels 7 (4) (2021), https://doi.org/10.3390/gels7040275.

[32] M.F. Ordoñez, G. Cerrato, A. Giordana, A. Di Michele, E. Falletta, C.L. Bianchi, One-pot synthesis of Ag-modified SrTiO3: synergistic effect of decoration and doping for highly efficient photocatalytic NOx degradation under LED, J. Environ. Chem. Eng. 11 (5) (2023), https://doi.org/10.1016/j.jece.2023.110368.
[33] P. Examiner, M. Kopec, 12, United States Patent 2 (12) (2010).
[34] J. He, X. Xu, M. Li, S. Zhou, W. Zhou, Recent advances in perovskite oxides for non-enzymatic electrochemical sensors: A review, Anal. Chim. Acta 1251 (2023), 341007, https://doi.org/10.1016/j.aca.2023.341007. September 2022.
[35] A.S. Patil, A.V. Patil, C.G. Dighavkar, V.A. Adole, U.J. Tupe, Synthesis techniques and applications of rare earth metal oxides semiconductors: A review, Chem. Phys. Lett. 796 (2022), 139555, https://doi.org/10.1016/j.cplett.2022.139555. February.
[36] Y. Zhang, L. Zhong, D. Duan, Single-step hydrothermal synthesis of strontium titanate nanoparticles from crystalline anatase titanium dioxide, Ceram. Int. 41 (10) (2015) 13516–13524, https://doi.org/10.1016/j.ceramint.2015.07.145.
[37] E. Grabowska, Selected perovskite oxides: Characterization, preparation and photocatalytic properties-A review, Appl. Catal. B Environ. 186 (2016) 97–126, https://doi.org/10.1016/j.apcatb.2015.12.035.
[38] R.I. Walton, Perovskite Oxides Prepared by Hydrothermal and Solvothermal Synthesis: A Review of Crystallisation, Chemistry, and Compositions, Chem. - A Eur. J. 26 (42) (2020) 9041–9069, https://doi.org/10.1002/chem.202000707.
[39] D.R. Modeshia, R.I. Walton, Solvothermal synthesis of perovskites and pyrochlores: Crystallisation of functional oxides under mild conditions, Chem. Soc. Rev. 39 (11) (2010) 4303–4325, https://doi.org/10.1039/b904702f.
[40] S.K. Gupta, Y. Mao, A review on molten salt synthesis of metal oxide nanomaterials: Status, opportunity, and challenge, Prog. Mater. Sci. 117 (2021), 100734, https://doi.org/10.1016/j.pmatsci.2020.100734. August 2020.
[41] P. Li, et al., Solvothermal synthesis and visible light-driven photocatalytic degradation for tetracycline of Fe-doped SrTiO3, RSC Adv 4 (88) (2014) 47615–47624, https://doi.org/10.1039/c4ra06630h.
[42] J.P. Zuniga, M. Abdou, S.K. Gupta, Y. Mao, Molten-salt synthesis of complex metal oxide nanoparticles, JoVE (Journal Vis. Exp. (140) (2018) e58482.
[43] H. Liu, X. Chen, S. Yan, Z. Li, Z. Zou, Basic molten salt route to prepare porous SrTiO3 nanocrystals for efficient photocatalytic hydrogen production, Eur. J. Inorg. Chem. (23) (2014) 3731–3735, https://doi.org/10.1002/ejic.201402280.
[44] GreenDelta, "LCIA methods: Impact assessment methods in life cycle assessment and their impact categories. Version 1.5.6," GreenDelta, no. 23, pp. 1–23, 2016, [Online]. Available: https://www.openlca.org/wp-content/uploads/2015/11/LCIA-METHODS-v.1.5.4.pdf.
[45] F. Gao, et al., Environmental impacts analysis of titanium sponge production using Kroll process in China, J. Clean. Prod. 174 (2018) 771–779, https://doi.org/10.1016/j.jclepro.2017.09.240.
[46] F. Yan, X. Zhang, E. Asselin, D. Duan, Z. Li, Preparation of strontium carbonate via celestite leaching in NaHCO3 using two interconnected reactors, Hydrometallurgy 204 (2021), 105729, https://doi.org/10.1016/j.hydromet.2021.105729. August.
[47] J.H. Lee, K.H. Ko, B.O. Park, Electrical and optical properties of ZnO transparent conducting films by the sol-gel method, J. Cryst. Growth 247 (1–2) (2003) 119–125, https://doi.org/10.1016/S0022-0248(02)01907-3.
[48] A. Kendall, Time-adjusted global warming potentials for LCA and carbon footprints, Int. J. Life Cycle Assess. 17 (8) (2012) 1042–1049, https://doi.org/10.1007/s11367-012-0436-5.
[49] E. Borsato, et al., Comparison of Water-focused Life Cycle Assessment and Water Footprint Assessment: The case of an Italian wine, Sci. Total Environ. 666 (2019) 1220–1231, https://doi.org/10.1016/j.scitotenv.2019.02.331.