



SrFeO₃ peculiarities and exploitation in decontamination processes and environmentally-friendly energy applications

Maria Laura Tummino

Institute of Intelligent Industrial Technologies and Systems for Advanced Manufacturing, Italian National Research Council, Corso G. Pella 16, 13900, Biella, Italy

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ABSTRACT

New technologies have been developed to avoid ecological damages by promoting depollution and eco-friendly synthesis routes of chemicals and materials, and setting efficient “cleaner” energy supply methods. These research fields, strongly interconnected, can contribute to sustainable technological advancement and represent the frame in which SrFeO₃-based materials can act as valuable tools. SrFeO₃ is a ABO₃ perovskite-type mixed oxide, widely studied in nanotechnology. Its pristine form does not contain expensive elements, such as rare and noble metals, or toxic components. SF peculiar features, such as non-stoichiometric composition (oxygen vacancies), the unusual oxidation state of Fe⁴⁺ and the related transformations of structural, electronic and magnetic characteristics, are attracting many research teams. This review summarizes most of these fundamental aspects, adding tips on some of the possible SF modification strategies and characterization techniques in view of environmental-benign applications. Particularly, thanks to SF-based materials’ oxygen exchange properties, ionic and electronic conduction abilities and catalytic activities, SFs are used in: (i) depollution processes as adsorbents, photo- and thermo-catalysts, combustion catalysts, antimicrobial agents, (ii) alternative devices for energy production and storage, and (iii) efficient systems for fuel conversion (i.e., chemical looping). The main correlations among SF composition, physical-chemical properties, performances and stability will be highlighted. This way, this review addresses indications to create SF-based tailored and functional materials specifically for the environment and energy, which are still underestimated and often replaced by other well-known oxides such as Lanthanum-based (in A-site) or Co, Ti-based (in B-site) perovskites.

1. Introduction

SrFeO₃ (Strontium Ferrate, SF) is a perovskite-type mixed oxide (PVO). Perovskites are described with the general formula (ABX₃), where A and B are metal cations and X is an anion, generally oxygen (ABO₃) [1,2]. The ideal cubic crystal structure of ABO₃-PVO consists of corner-sharing [BO₆] octahedra with the A cation occupying the 12-fold coordination site formed in the middle of the cube surrounded by eight such octahedra. In particular, the A-site is occupied by alkali, alkaline-earth and rare-earth metals whereas transition metals cations generally occupy B-site. The ideal cubic perovskite is not very common and can evolve in tetragonal, rhombohedral or orthorhombic structures, depending on the distortion degree. To describe the stability of PVOs, Goldschmidt proposed the tolerance factor “*t*”, a geometrical parameter defined based on ionic radii [1,3,4]. A *t* value close to 1 is found for ideal cubic or slightly distorted structures, whereas lower/higher *t* values are typical of strongly distorted ones; *t* values

too far from 1 indicate perovskite-type structures with insufficient stability.

Among various PVOs, SF is advantageous since, in its basic form, it has low toxicity and is free from rare-earth elements and precious metals [5,6], also reducing production costs. Moreover, SF possesses particular physical-chemical features that confer specific activities to this oxide, which recently became ever more attractive to the scientific community, in different technological fields, including environmental preservation and clean energy production methods, as shown hereinafter.

2. SrFeO₃ main features, synthetic strategies and characterization methods

In SrFeO₃ (Fig. 1a), iron has the unusual oxidation state of +4 [3,4], whereas, in most ABO₃ perovskites, the B-site oxidation state is +3. Depending on the temperature and partial pressure conditions, SF can accommodate an extensive range of oxygen deficiencies (resulting in a

E-mail address: marialaura.tummino@stiima.cnr.it.

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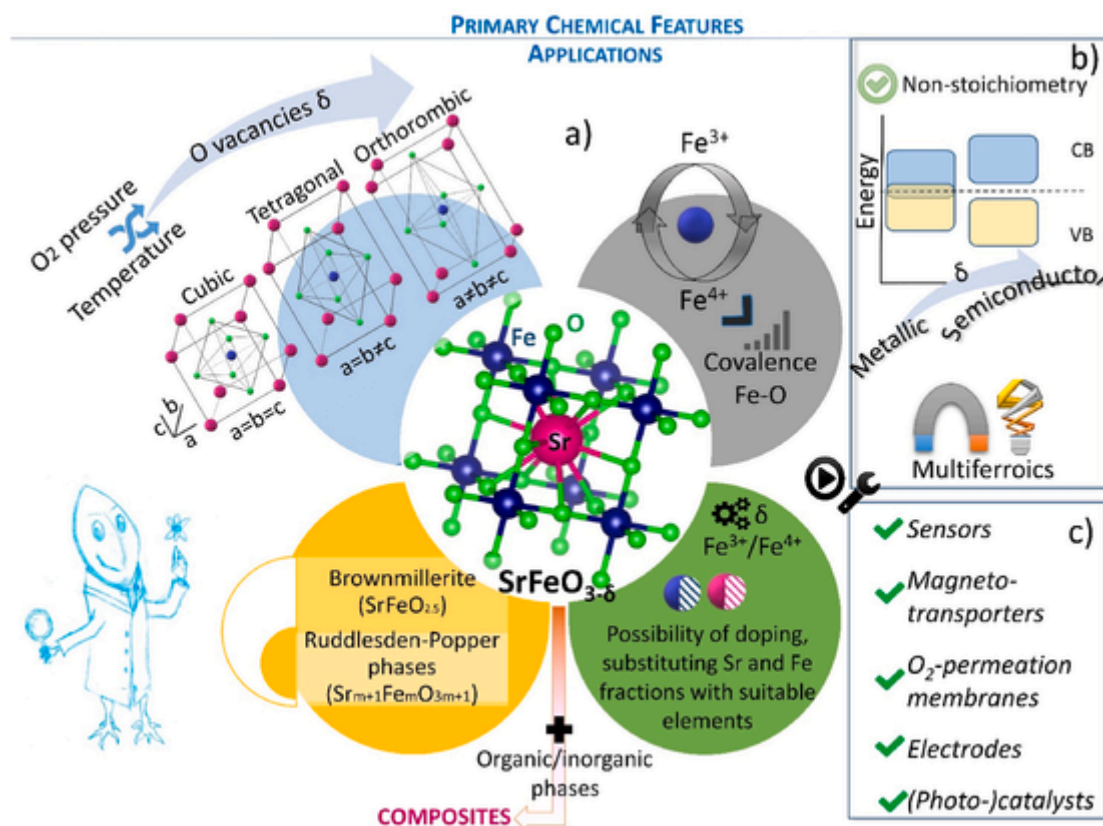


Fig. 1. Structure and primary information on SrFeO_3 perovskite-type oxides; (a) focus on: the chemical features related to the microstructure, - the presence of the oxygen vacancies and the unusual valence state of iron IV and their relationships with the crystalline structure, - the possibility to dope the materials in both A and B sites varying the non-stoichiometry and tuning the properties; (b) the influence of non-stoichiometry on the transition from metallic to semiconducting behavior of SrFeO_3 , also characterized by multiferroic effects; (c) list of some of the main applications of SrFeO_3 -based compounds, which can also be combined in composite materials.

general formula $\text{SrFeO}_{3-\delta}$, where δ stands for oxygen non-stoichiometry value), thus some Fe^{3+} , with a larger ionic radius, is formed to balance the global neutral valence state, creating the redox couple $\text{Fe}^{4+}/\text{Fe}^{3+}$ [3,7]. Doped SF can be described by the formula $\text{Sr}_{(1-x)}\text{A}'_x\text{Fe}_{(1-y)}\text{B}'_y\text{O}_{3-\delta}$ [7], where A' , B' are the dopants and x , y are the substituting fractions of the primal elements. A' and B' can be any metal cations selected to maintain the overall steric characteristics and a t -factor not too far from unity. After introducing doping elements with different oxidation states, a new electroneutrality condition is restored by cations' partial oxidation or reduction and the eventual formation of new oxygen vacancies [3]. Connected to these aspects, SFs undergo crystal structural changes with the δ values [8]: SFs are cubic at $\delta = 0$, tetragonal at $\delta = 0.125$, orthorhombic at $\delta = 0.25$, brownmillerite-type (a vacancy-ordered phase) at $\delta = 0.5$ and have mixed phases in between δ values. It is possible to obtain also Ruddlesden–Popper-type (RP) SFs, consisting of an alternating arrangement of SrFeO_3 layers sandwiching a rock salt layer (SrO) [9]. Such oxygen non-stoichiometry and structural versatility are essential factors in achieving tunable oxygen adsorption, oxygen mobility, and ionic conductivity considering the applications (an exemplifying list in Fig. 1c [8,10]). For instance, the presence of Fe^{4+} cations within a cubic symmetry, enhancing Fe–O covalency, maximizes the overlapping and the charge transfer between the $2p$ -orbitals of oxygen and d -orbitals of the B-site transition metals [3,11]. Moreover, as summarized in Fig. 1b, SF electrical conductivity ranges from a metallic-like behavior (in cubic phases with Fe^{4+}), to semiconducting behavior (for oxygen-deficient SF) [8]. Depending on δ , SF exhibited both antiferromagnetism and the multiferroic effect, a phenomenon in which a material shows at least two ferroelectric parameters (ferromagnetism, ferroelectricity, or ferroelasticity), widening the application fields of SF in nanoelectronics [4,8,11].

As for other PVOs, SF physicochemical properties can be influenced by synthesis methods and their intrinsic parameters (i.e., metal precursors, reactants, operating temperature and pressure, physical-chemical environments, formation of composites, etc.) [3,12–15]. Some of the most used methods are represented in Fig. 2 and briefly described herein below:

- (1) solid-state reaction: mechanical mix of precursors followed by high-temperature treatment;
- (2) co-precipitation reaction: mixing of soluble metal precursors in supersaturation conditions with a precipitation agent;
- (3) hydrothermal synthesis: a crystallization carried out in autoclave at high temperatures and high pressures;
- (4) spray pyrolysis: a solution of metal precursors nebulized into an oxy-hydrogen flame forming nanoparticles;
- (5) sol-gel synthesis: evolution of a colloidal solution into a gel-like network followed by drying and calcination;
- (6) solution combustion synthesis: a modified sol-gel procedure in which a thermally-induced and self-sustaining redox reaction takes place between oxidant metal precursors and reducing agents.

Considering these methods, some of them require high energetic conditions, such as (1), (3), (4), whereas others need a careful and simultaneous regulation of temperature, pH, reactant concentration and solubility, such as (2), (5), (6) [16]. The physical-chemical characteristics mostly affected by synthesis are related to the phase purity and homogeneity, the (micro-)structure (crystallinity, particle size and distribution) and surface morphology. Among all, sol-gel and solution combustion synthesis have received great attention since they are low-

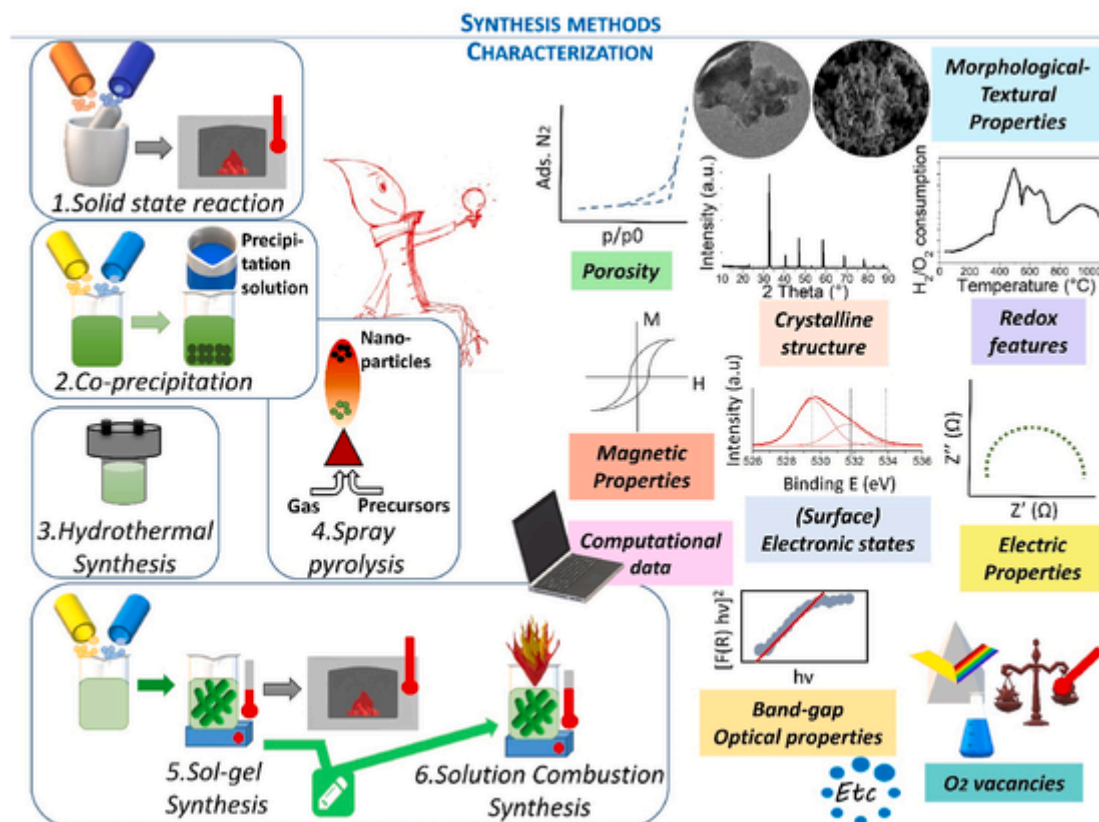


Fig. 2. On the left, representations of the most employed synthesis methods: (1) solid-state reaction, (2) co-precipitation reaction, (3) hydrothermal synthesis, (4) spray pyrolysis, (5) sol-gel synthesis, (6) solution combustion synthesis. On the right, properties to be examined in relation to the applications, such as porosity and surface area (by gas adsorption techniques), texture and morphology (by microscopies), crystalline structure (by X-ray diffraction), redox behavior (by Temperature Programmed Analyses), magnetic properties (by magnetometry), electronic features (by photoemission and photoabsorption spectroscopy), electric properties (by impedance spectroscopy and voltammetry), optical features (by UV-vis spectroscopy), O-vacancies determination (by spectroscopical/thermal/titration methods). Computational studies can also be exploited to deepen these aspects.

temperature and low-cost methods; they also allow better control over the structural and microstructural features and a high dopant concentration [17].

This wide variety of tunable features needs specific characterization techniques able to explain and, possibly, predict the performances of SFs. Fig. 2 displays a non-exhaustive list of characterizations: indeed, other methods exist for SF investigations and some of the reported ones can be used for more than one aspect.

3. Decontaminating activities of SrFeO₃

A traditional depuration method is pollutant adsorption (Fig. 3a), namely the action of solids to attract to their surface molecules from gases or solutions they are in contact with, by either physical or chemical interactions. Generally, low Specific Surface Area (SSA) values and porosities characterize SFs, but preparation routes can improve them (see mechanical grinding, kinetics of the synthetic reaction, sintering temperature, combination with high-surface compounds) [3,10,11,13,14,18,19]. Similarly, the surface charge of PVOs is tunable and influences the electrostatic affinity adsorbent/adsorbate [10,19,20]. SF-based materials adsorption capabilities in water remediation have been reported towards dyes, oil residues, chloramphenicol and bisphenol A [10,18,19,21]. Beyond being a self-sufficient method, adsorption is determining for further treatments requiring interfacial contact, such as catalytic processes [10,14,19,21].

Actually, with band-gap energy values between 1.80 and 3.75 eV, SF compounds found a place as photocatalysts irradiated by UV and visible light for aqueous pollutant removal (Fig. 3b) [10]. Doped and undoped SFs induced the degradation of nitrobenzene, phenol and differ-

ently-charged dyes, due to the light-driven generation of ROS (Reactive Oxygen Species) as hydroxyl radical, superoxide anion radical and singlet oxygen, whose formation is favored by the O-mobility and the redox couple Fe³⁺/Fe⁴⁺ [10,19]. SF was also employed in heterojunctions. SrFeO_{3-δ}/graphitic-C₃N₄ with hierarchical architecture showed enhanced visible-light photodegradation of chloramphenicol and crystal violet, due to the suppressed recombination of electron-hole pairs, and also an easy recoverability taking advantage of the magnetic properties of SF [18]. Another study pointed out the convenience of using SF in combination with TiO₂, thanks to SF's ability to absorb light and its cubic organization that exhibits the greatest degree of electronic guiding, thus increasing the separation efficiency of photogenerated carrier pairs [13].

SF activity was also ascertained in dark ambient degradation experiments, which require simple and low-energetic conditions. Bisphenol A and acid orange 8 were efficiently adsorbed and then mineralized onto a positively-charged ball-milled SF surface, rich in O₂^{•-} [19]. Additionally, A and B site-substituted SFs have been proposed to initiate the dark degradation of ofloxacin through peroxymonosulfate activation, governed by ROS evolution [22]. In both works, enhanced superficial δ powered the SF activity.

A Sr_{0.85}Ce_{0.15}FeO_{3-δ} (SCF), initially studied as a photocatalyst, proved to be sensitive to mild temperatures in a dark environment (Fig. 3c). Within a narrow temperature range (55–80 °C), SCF performed efficiently in Orange II abatement, due to the temperature-dependent increment of •OH generation [10]. This first work was followed by another research, in which SCF was used, without any additional energy input, to degrade oil residues in the so-called “produced water” that commonly comes out at 40–50 °C [21]. After initial batch tests, a lab-

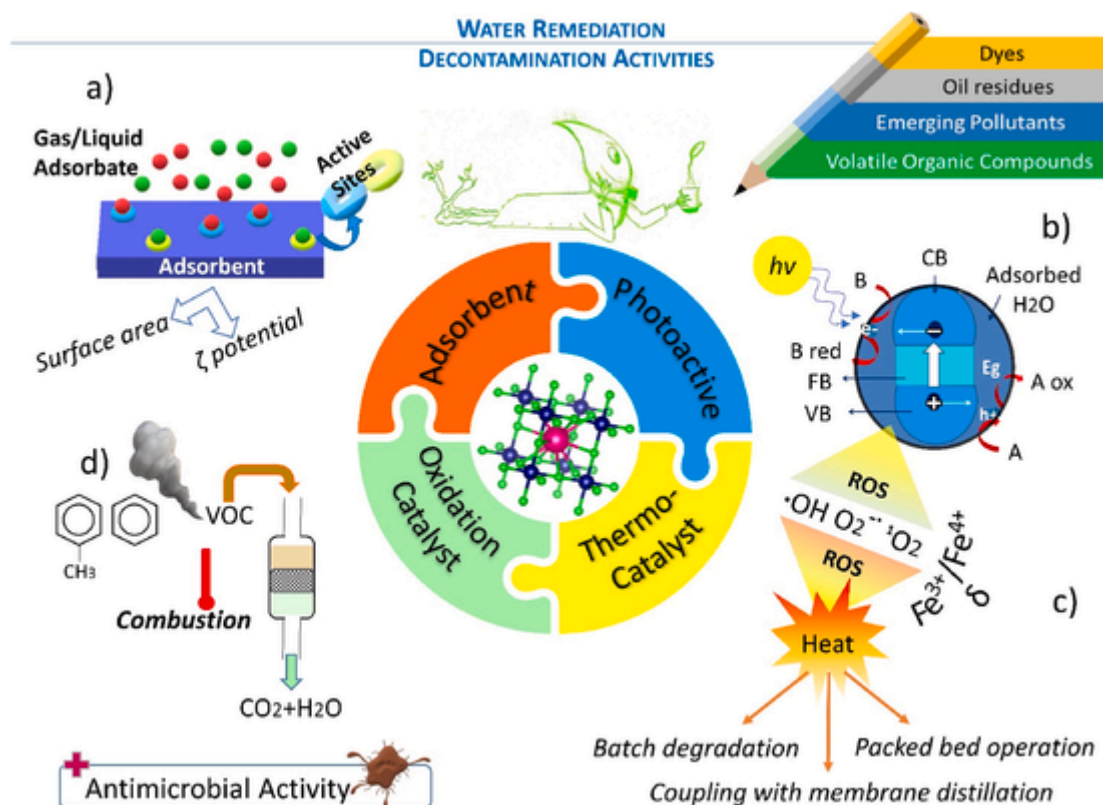


Fig. 3. Environmental activities of SrFeO_3 perovskite-type oxides for various pollutant removal, related to (a) adsorption properties led by surface area properties and surface charge (determined, for instance, by zeta potential measures); (b) photocatalysis (the scheme represents a semiconductor photocatalyst based on the electron/hole pair excitation causing redox reactions) driven by the generation of Reactive Oxygen Species (ROS), also correlated to oxygen vacancies and the duality among Fe valence states, (c) thermocatalytic activity, with similar mechanisms as (b), after subjection to mild temperature and already tested in the literature in different reaction conditions; (d) catalytic oxidizing action towards Volatile Organic Compounds (VOC) in a sketched reactor. In addition, antimicrobial activity was ascertained towards Gram-positive, Gram-negative bacteria and fungi strains. In (b), the acronyms are VB = valence band, CB = conduction band, FB = Forbidden Band, Eg = Energy gap.

scale packed bed reactor was prepared for continuous experiments, showing the future potentialities of SCFs in offshore treatments of produced water. Moreover, Janowska et al. [23] proposed an integrated process in which thermal energy simultaneously drove the permeation of pure water through a hydrophobic membrane (membrane distillation) and activated the thermocatalytic SCF for bisphenol A degradation.

Fig. 3d displays Strontium Ferrates as oxidation catalysts for VOCs, such as toluene and benzene (aromatic compounds affecting air quality, with a low reactivity for combustion, that need catalytic processes) [5,7]. One of the first approaches concerned 3D-ordered macroporous cubic SFs, prepared using a templating method: their efficiency in toluene combustion ($T_{90\%}$ between 340 and 492 °C) was connected to high SSA (derived by the synthesis method), O_{ads} species concentration, and SF low-temperature reducibility [14]. Analogous effects were observed by Hashimoto et al. [5], who prepared a series of $\text{SrFe}_{1-y}\text{Sn}_y\text{O}_{3-\delta}$. The Sn-doping caused the SSA increase, resulting in the acceleration of Fe redox rates at 500 °C and higher catalytic activity in benzene combustion.

Lastly, SF has been recognized as a broad-spectrum antimicrobial agent [11]: the action mechanism was ascribable to nano-size, Sr^{2+} liberation, Fe^{3+} , $\text{Fe}^{3.5+}$, and Fe^{4+} states, and the oxygen deficiency that enhanced surface reactivity.

4. Use of SrFeO_3 in green energy production and storage

Given the constant increment of global energy demand and the linked environmental issues, novel methods of clean-burning fossil fuel, the use of renewable sources and the development of alternative sys-

tems enabling a greener conversion between electrical and chemical energies have drawn significant attention. Furthermore, most renewables (that, contrarily to fossil fuels, can neither be kept in physical containers nor transported) need to be first converted to electricity, which, in turn, must be stored in proper devices. $\text{SrFeO}_{3-\delta}$ possesses suitable features to be employed in these technologies, as elucidated in Fig. 4.

For example, SFs are employable as cathodes and anodes (due to the bivalent reductive-oxidizing activities) for solid oxide electrochemical cells (Fig. 4a), recognized as clean-energy devices. In SOFC mode, several fuels, including H_2 and hydrocarbons, can be oxidized to produce electricity, generally at elevated temperatures, although further progress has been made to moderate the operating conditions (500–800 °C in IT-SOFCs) [3,24]. Conversely, in SOECs, electrical energy is supplied to drive the reversed reactions (i.e., obtaining H_2 from H_2O) [25]. For these scopes, A-site doping (with Ce, Sm, La) is recommended to stabilize the catalytically favorable cubic structure and further decrease TEC, together with the B-site substitution with elements able to improve structural stability, MIEC and overall performances (such as Co, Mo, Ti, Nb, Zr, Cu, W, Ru, Cr, Sb) [3,15,24,25]. Also, the synthesis methods, influencing (micro-)structural properties, may affect the electrochemical properties [3,16,17].

Similarly, SFs have been proposed as cathodes in rechargeable metal-air batteries [26,27] (Fig. 4b), involved in both Oxygen Evolution Reaction and Oxygen Reduction Reaction (OER/ORR [12]), for instance, in a Mg-battery [27]. An interesting case concerns a doped-RP $\text{LaSr}_3\text{Co}_m\text{Fe}_{3-m}\text{O}_{10-\delta}$ bifunctional material that exhibited improved stability and performances due to enhanced δ , facile oxygen exchange, synergistic interplay Co/Fe and a narrowed potential-gap [26]. Also, to support renewables such as solar energy (a time-variable source), TCES

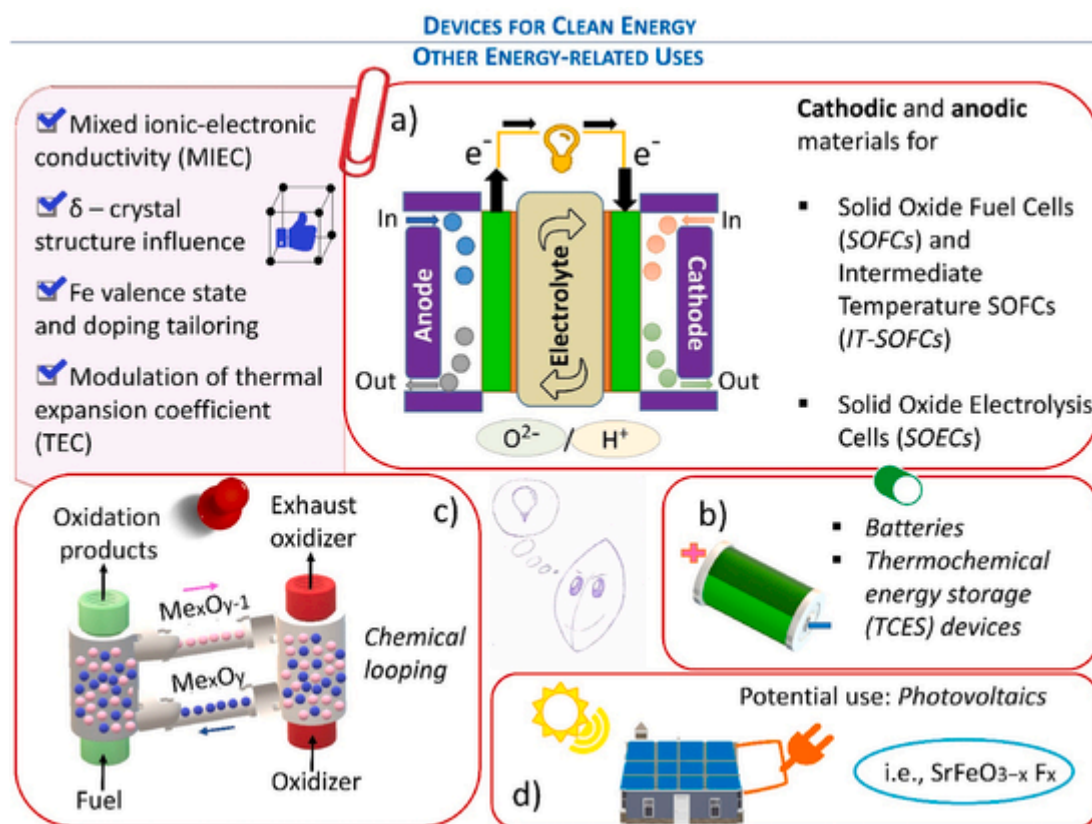


Fig. 4. SrFeO_3 perovskite-type oxides as suitable materials in energy-related applications as (a) electrodes for different fuel cells (the type of device regulates the transported ion species, either O^{2-} or H^+), being capable of adsorbing oxygen and catalytically reducing it or evolving oxygen and catalytically oxidize the fuel, and possessing high MIEC and TEC compatibility with the electrolyte to avoid detrimental effects on the performances and failures in long-term operations; (b) electrodes for energy storage devices, exploiting similar properties as in (a); (c) solid oxygen carriers in chemical looping systems, which provide more efficient fuel conversion with an intrinsic capability of highly efficient CO_2 separation; (d) possible optical-absorber in photovoltaics (the example reports fluorine-substituted $\text{SrFeO}_{3-x}\text{F}_x$ epitaxial thin film).

systems can ensure the continuity of energy supply [28,29]. In TCES, thermal energy drives a reversible endothermic chemical reaction, storing the energy as chemical potential. Darwish et al. investigated under redox cycling the oxygen release/consumption behavior and reaction enthalpies of Mn- or Cu-substituted SrFeO_3 for thermochemical applications [28]. Copper significantly increased the cyclic stability and oxygen exchange capacity [28], but the careful regulation of Cu amount has been assessed as an essential parameter [29].

Chemical looping offers a potentially efficient and cost-effective approach for carbonaceous energy conversion, CO_2 capture, and chemical production (see syngas), with different configurations and chemical feeds, i.e., methane, ethane, ethylene. In chemical looping, an oxygen carrier spontaneously releases and replenishes lattice oxygen under oxygen partial pressure and/or temperature swings (scheme in Fig. 4c) [6,9,30,31]. Several doped SF sorbents, such as $\text{Sr}_x\text{Ce}_{1-x}\text{FeO}_{3-\delta}$, $\text{Sr}_x\text{Ca}_{1-x}\text{FeO}_{3-\delta}$, $\text{SrFe}_y\text{Cu}_{1-y}\text{O}_{3-\delta}$, $\text{SrFe}_y\text{Co}_{1-y}\text{O}_{3-\delta}$ and $\text{SrFe}_y\text{Mn}_{1-y}\text{O}_{3-\delta}$, have demonstrated suitable thermodynamic properties, excellent redox stability and oxygen-carrying capacity at relatively low temperatures.

Regarding the potentialities of SF in photovoltaic applications (Fig. 4d), often appanage to organic–inorganic halide perovskites, Katayama et al. [32] were inspired by the substitution of F^- as an effective approach to alter the electric and magnetic properties of iron PVOs. $\text{SrFeO}_{2-x}\text{F}_x$ epitaxial thin films have been fabricated by a topotactic fluorination technique with a sufficiently high crystalline quality for electronic and optical measurements. In addition, the controllability of fluorine content would enable band-gap engineering, which is of significant importance for tailoring the optical properties and electronic band structure of photovoltaic materials.

5. Challenges and final remarks

Beyond the specific activities of SFs, other operating aspects must be heard in mind.

For instance, as anticipated in Fig. 4 and its caption, significant discrepancies among TEC values between electrodes and electrolytes in electrochemical cells can lead to a mismatch between the components and consequent thermomechanical instability. This phenomenon can be avoided by carefully choosing A- and B-site dopants in SF electrocatalysts [3,15,24].

More generally, the possibility to use SFs over several cycles is a discriminating factor for the applicability of these compounds, both as catalysts and oxygen exchangers/carriers. This requirement is accomplished when the materials show resistance and stability to the operating conditions without the inactivation of active sites and/or when they can be regenerated, as already established for SFs and doped SFs [6,15,28].

In fact, the poisoning of the active materials is an issue in many processes. For instance, doped SFs have been proven resistant to poisoning derived from reactants or their by-products involved during the operations, as reported in Ref. [24]. Another source of poisoning is related to the formation of SrCO_3 due to the SF interaction with CO_2 [2,10]. Østergaard et al. [2] have recently pointed out that in SCF thermocatalyst designed for wastewater depuration, the carbonate formation occurred when the catalyst was dispersed in water and, faster, when it was used in the degradation of bisphenol A (reducing its catalytic efficiency). In contrast, no SrCO_3 was detected for samples stored in air for up to 195 days. In any case, carbonate formation is a ther-

mally-reversible process, and SCF could resume its activity after heat treatment at 900 °C for 1 h [2].

In summary, this review would give food for thought regarding SrFeO_{3-δ} perovskites as promising tools in sustainable chemistry, due to their remarkable stability and versatility. Thanks to peculiar structural-compositional features, especially O-vacancy accommodation and Fe oxidation states, SFs offer broader possibilities to be finely-tuned (i.e., modifying preparation conditions and doping elements) and exploited for ecological applications in the interrelated fields of environmental remediation and sustainable energy supply.

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Maria Laura Tummino : Conceptualization, Methodology, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The author declares that she has 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.

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