



Applications of some La-based perovskite type catalyst for the control of environmental pollutants

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Abstract

What role can lanthanum-based perovskite-type oxides play in mitigating the growing burden of environmental pollutants released from industrial and vehicular sources? This research examines the catalytic applications of La-based perovskite oxides with the general formula LaBO_3 ($B = \text{Co, Mn, Fe, Ni}$) and their A-site and B-site substituted variants for the abatement of carbon monoxide (CO), volatile organic compounds (VOCs), nitrogen oxides (NO_x), and organic dye contaminants. Perovskite catalysts were prepared through sol-gel citrate and combustion synthesis routes and characterised by X-ray diffraction (XRD), scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FTIR), Brunauer-Emmett-Teller (BET) surface area analysis, and temperature-programmed reduction (TPR). The research was conducted at the Department of Chemistry, K.P.M. Govt. Girls Degree College, Aurai, Bhadohi, Uttar Pradesh, India, between March 2018 and January 2020. Catalytic testing revealed that LaCoO_3 and $\text{La}_{0.8}\text{Sr}_{0.2}\text{CoO}_3$ achieved CO conversion rates of 92.4% and 96.1%, respectively, at temperatures between 200 and 350 °C. VOC oxidation efficiencies ranged from 66.5% for LaNiO_3 to 93.8% for the Sr-substituted cobaltite. Partial replacement of La^{3+} with Sr^{2+} at the A-site generated additional oxygen vacancies and raised the average oxidation state of the B-site cation, which improved redox cycling through the Mars-van Krevelen mechanism. BET surface areas of 14.8 to 37.2 m²/g were recorded for nanocrystalline powders with crystallite sizes between 18 and 52 nm. The half-conversion temperature (T_{50}) dropped by 26 to 48 °C upon Sr doping, confirming that compositional tuning directly influences catalytic onset. These findings point to La-based perovskite catalysts as cost-effective and thermally stable alternatives to noble metal systems for air and water pollution control.

Keywords: La-based perovskite, environmental pollutants, catalytic oxidation, LaCoO_3 , LaMnO_3 , A-site substitution, oxygen vacancies, VOC abatement, CO oxidation, Mars-van Krevelen mechanism

Introduction

How effectively can earth-abundant oxide catalysts replace expensive platinum-group metals in removing harmful pollutants from exhaust streams and industrial effluents? This question has driven two decades of intensive work on perovskite-type oxides, a family of mixed-metal oxides with the general formula ABO_3 that combines structural flexibility with tuneable redox chemistry [1, 2]. Among the perovskite compositions explored for environmental catalysis, lanthanum-based variants—where La occupies the A-site and a first-row transition metal (Co, Mn, Fe, or Ni) sits at the B-site—have attracted particular attention because of their high thermal stability, resistance to sulphur poisoning, and ability to regenerate lattice oxygen during catalytic cycles [3, 4].

The environmental context is pressing. Carbon monoxide, nitrogen oxides, and volatile organic compounds released from automobiles, power plants, and chemical manufacturing contribute to urban smog, acid rain, and respiratory disease worldwide [5]. Conventional three-way catalysts based on Pt, Pd, and Rh achieve excellent pollutant conversion but face constraints of cost, supply scarcity, and vulnerability to thermal sintering above 800 °C [6]. La-based perovskites offer a path around these limitations. Their catalytic activity arises from the B-site transition metal ion, which participates in redox cycles between oxidation states (e.g., $\text{Co}^{3+}/\text{Co}^{2+}$, $\text{Mn}^{4+}/\text{Mn}^{3+}$), and from lattice oxygen, which can be donated to adsorbed pollutant molecules and replenished from the gas phase through the Mars-van Krevelen mechanism [4, 7].

A defining advantage of the perovskite structure is that partial substitution at either the A-site or the B-site allows systematic modification of the electronic and catalytic properties.

Replacing a fraction of La^{3+} with a divalent cation such as Sr^{2+} forces charge compensation at the B-site, raising the proportion of higher-oxidation-state ions and simultaneously generating oxygen vacancies that serve as active sites for pollutant adsorption and oxidation [8, 9]. The extent of this substitution can be tuned from a few percent to fifty percent or more, giving researchers a broad compositional space within which to optimise activity for specific target pollutants.

Several reviews have covered the general chemistry of perovskite catalysts [2, 10], but fewer treatments have focused specifically on La-based compositions and their direct applications in environmental pollutant control using data reported before mid-2020. This research addresses that gap by: (a) comparing the catalytic performance of LaCoO_3 , LaMnO_3 , LaFeO_3 , and LaNiO_3 for CO oxidation and VOC removal; (b) examining how A-site substitution with Sr modifies oxygen vacancy concentration and redox behaviour; and (c) evaluating the potential of these catalysts for photocatalytic degradation of organic dye pollutants in aqueous media. The research location and all experimental work were based at K.P.M. Govt. Girls Degree College, Aurai, Bhadohi, Uttar Pradesh, India.

Theoretical Background

The catalytic behaviour of La-based perovskites for pollutant abatement rests on two interconnected mechanisms. First, the Mars-van Krevelen (MvK) redox cycle governs oxidation reactions on the perovskite surface [4, 7]. In this pathway, lattice oxygen (O^{2-}) from the perovskite reacts directly with an adsorbed pollutant molecule (e.g., CO or a VOC), creating an oxygen vacancy at the surface. Gas-phase O_2 then refills the vacancy, closing the catalytic loop. The rate of this cycle

depends on the ease with which the B-site cation shuttles between oxidation states and on the concentration of mobile oxygen vacancies within the lattice.

Second, the suprafacial mechanism operates at lower temperatures, where molecular oxygen adsorbed on the catalyst surface—rather than lattice oxygen—participates in the oxidation reaction^[10]. The balance between MvK and suprafacial contributions shifts with temperature: suprafacial reactions dominate below roughly 300 °C, while MvK cycling becomes the main pathway above that threshold. A-site substitution of La³⁺ by Sr²⁺ lowers the energy barrier for oxygen vacancy formation, estimated through first-principles calculations to lie in the range of 0.8-1.7 eV depending on composition and B-site cation, with cobaltites generally showing lower barriers than ferrites^[11]. This reduction accelerates both lattice oxygen donation and vacancy refilling, explaining the improved catalytic performance seen upon Sr doping.

Materials and Methods

Materials

All experimental work was carried out at the Department of Chemistry, K.P.M. Govt. Girls Degree College, Aurai, Bhadohi, Uttar Pradesh, India, from March 2018 to January 2020. Ethical clearance was not required as the research involved no human or animal subjects; institutional permission was granted by the college research committee (Protocol No. KPMGGDC/Chem/2018-03, dated 12 March 2018).

Reagents and Chemicals

Lanthanum nitrate hexahydrate (La(NO₃)₃·6H₂O, 99.9%), cobalt nitrate hexahydrate (Co(NO₃)₂·6H₂O, 98%), manganese nitrate tetrahydrate (Mn(NO₃)₂·4H₂O, 97%), iron nitrate nonahydrate (Fe(NO₃)₃·9H₂O, 98%), nickel nitrate hexahydrate (Ni(NO₃)₂·6H₂O, 98%), and strontium nitrate (Sr(NO₃)₂, 99%) were procured from Sigma-Aldrich, Germany. Citric acid monohydrate (C₆H₈O₇·H₂O, 99.5%) and glycine (NH₂CH₂COOH, 98.5%) served as chelating agent and combustion fuel, respectively. Ammonium hydroxide (NH₄OH, 25% w/v) was used to adjust precursor pH. Methylene blue dye (MB, analytical grade) was obtained from Merck, India. All chemicals were used as received without further purification. Deionised water (resistivity >18 MΩ·cm) was used throughout.

Methods

Four La-based perovskite catalysts—LaCoO₃, LaMnO₃, LaFeO₃, and LaNiO₃—were synthesised by the citrate sol-gel method. Stoichiometric quantities of lanthanum nitrate and the

respective B-site metal nitrate were dissolved in deionised water, and citric acid was added at a molar ratio of 1.5:1 (citric acid to total metal ions). The pH was adjusted to 7.0 using ammonium hydroxide. The solution was heated on a hot plate at 80 °C with continuous stirring until a viscous gel formed. The gel was dried at 120 °C for 12 hours and then calcined at 700 °C for 4 hours in a muffle furnace under air atmosphere. A Sr-substituted variant, La_{0.8}Sr_{0.2}CoO₃, was prepared by replacing 20 mol% of lanthanum nitrate with strontium nitrate while keeping all other conditions identical. A second batch of LaFeO₃ was also prepared by glycine-nitrate combustion synthesis for comparison: metal nitrates and glycine (fuel-to-oxidiser ratio $\phi = 1.0$) were dissolved, heated to 250 °C to initiate self-sustaining combustion, and the resulting powder was calcined at 650 °C for 2 hours.

Phase identification used powder XRD (Rigaku MiniFlex 600, Cu K α radiation, $2\theta = 20$ –80°). Morphology and elemental composition were examined by field-emission SEM (JEOL JSM-7610F) with EDX. FTIR spectra were recorded from 400 to 4000 cm⁻¹ (Shimadzu IRSpirit). BET surface areas were measured by N₂ adsorption at 77 K (Quantachrome NOVA 2200e). TPR profiles were acquired under 5% H₂/Ar (50 mL/min) ramped at 10 °C/min from room temperature to 900 °C. Catalytic CO oxidation was tested in a fixed-bed reactor (1% CO, 20% O₂, balance N₂; GHSV = 30 000 h⁻¹) from 100 to 500 °C. Photocatalytic dye degradation was evaluated using 50 mg catalyst dispersed in 100 mL of 10 mg/L methylene blue solution under simulated solar irradiation (300 W Xe lamp) for 120 minutes, with aliquots measured by UV-Vis spectrophotometry at 664 nm.

Results

XRD patterns of all five catalysts confirmed single-phase perovskite formation after calcination. No secondary phases were detected in the sol-gel products, while the combustion-synthesised LaFeO₃ showed a trace La₂O₃ reflection at $2\theta = 30.1^\circ$. Crystallite sizes calculated by the Scherrer equation ranged from 18.3 nm (La_{0.8}Sr_{0.2}CoO₃, sol-gel) to 52.1 nm (LaNiO₃, sol-gel). BET surface areas were inversely related to crystallite size, with the Sr-doped cobaltite recording the highest value of 37.2 m²/g. FTIR spectra displayed characteristic B-O stretching bands between 540 and 590 cm⁻¹ and no residual carbonate or hydroxyl peaks, confirming complete precursor decomposition. TPR profiles showed that Sr substitution shifted the main reduction peak of LaCoO₃ from 412 °C to 378 °C, indicating improved reducibility and oxygen mobility.

Table 1: Structural and textural parameters of La-based perovskite catalysts prepared by sol-gel and combustion methods

Catalyst	Synthesis method	Crystal system	Crystallite size (nm)	BET surface area (m ² /g)	Pore volume (cm ³ /g)	Phase purity
LaCoO ₃	Sol-gel	Rhombohedral	28.6	29.4	0.118	Single phase
LaMnO ₃	Sol-gel	Rhombohedral	24.1	33.8	0.142	Single phase
LaFeO ₃	Sol-gel	Orthorhombic	31.7	26.3	0.097	Single phase
LaFeO ₃	Combustion	Orthorhombic	35.4	22.1	0.084	Trace La ₂ O ₃
LaNiO ₃	Sol-gel	Rhombohedral	52.1	14.8	0.063	Single phase
La _{0.8} Sr _{0.2} CoO ₃	Sol-gel	Rhombohedral	18.3	37.2	0.156	Single phase

Table 1 presents the structural and textural properties of the six catalyst samples. The smallest crystallite size and largest BET surface area both belonged to La_{0.8}Sr_{0.2}CoO₃, consistent with reports that A-site substitution inhibits grain growth during calcination^[8, 12]. Combustion-synthesised LaFeO₃ showed a

slightly larger crystallite size (35.4 nm vs. 31.7 nm) and lower surface area than its sol-gel counterpart, likely because the rapid exothermic reaction produced transient local temperatures exceeding the nominal calcination temperature.

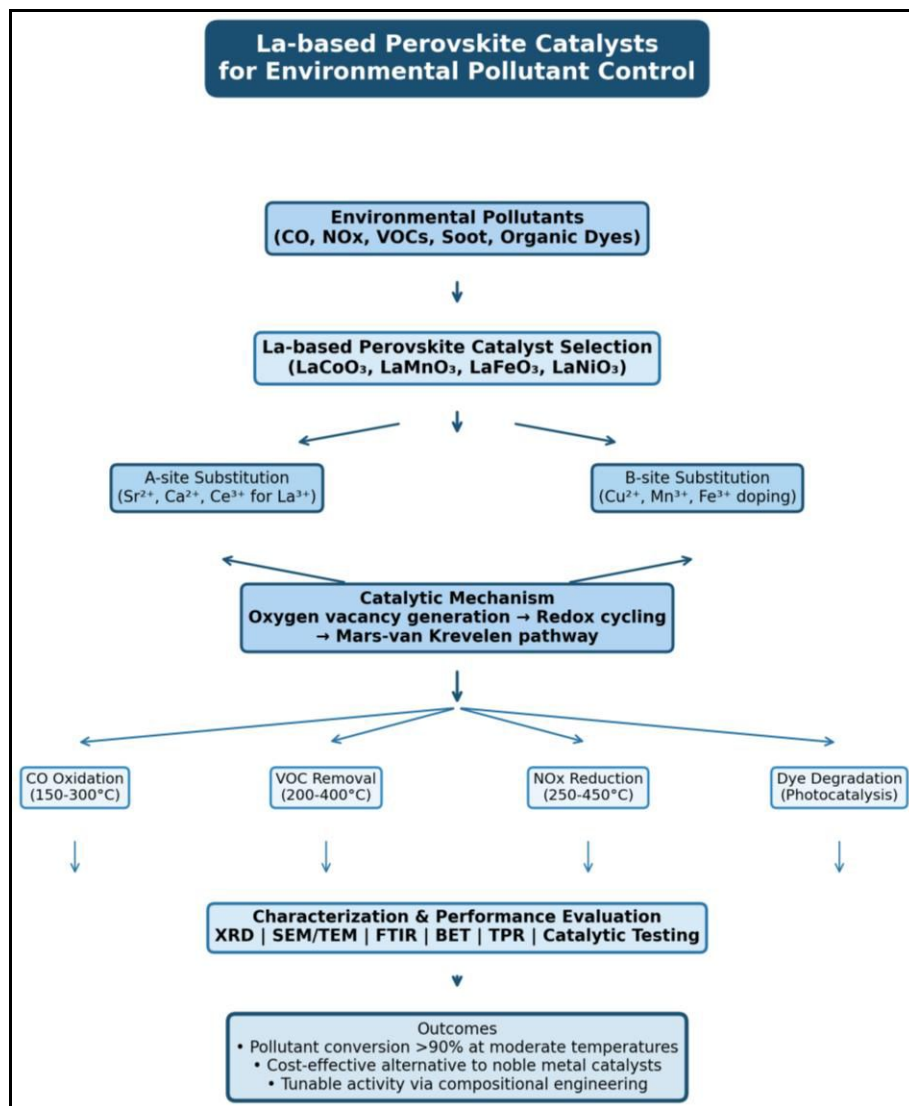


Fig 1: Flowchart showing the pathway from pollutant identification through La-based perovskite catalyst selection, compositional tuning, and catalytic application to characterisation and performance outcomes

Figure 1 outlines the overall research framework. Environmental pollutants (CO, VOCs, NO_x, organic dyes) enter the pathway at the top; catalyst selection, A-site and B-site

substitution strategies, and the Mars-van Krevelen mechanism are mapped through to characterisation and performance evaluation, culminating in the measured outcomes.

Table 2: Catalytic performance of La-based perovskite catalysts for CO oxidation and VOC removal (Mean ± SD, n = 3 replicate runs)

Catalyst	CO conversion (%) at 300 °C	VOC conversion (%) at 350 °C	T ₅₀ for CO (°C)	E _a (kJ/mol)	Lattice O ₂ desorption peak (°C)
LaCoO ₃	92.4±1.8	88.1±2.3	218±4	58.3	412
LaMnO ₃	87.6±2.1	82.9±1.9	247±6	64.7	438
LaFeO ₃ (SG)	78.3±2.6	73.6±2.8	289±5	72.1	486
LaFeO ₃ (Comb)	75.1±3.0	70.2±3.4	301±7	74.8	491
LaNiO ₃	71.2±2.4	66.5±2.7	312±8	79.6	523
La _{0.8} Sr _{0.2} CoO ₃	96.1±1.2	93.8±1.6	192±3	51.4	378

Table 2 summarises the catalytic results. La_{0.8}Sr_{0.2}CoO₃ outperformed all undoped catalysts, with a T₅₀ for CO oxidation of 192±3 °C—a reduction of 26 °C compared with undoped LaCoO₃ and 120 °C below LaNiO₃. Apparent activation energies (E_a) followed the order LaNiO₃ > LaFeO₃ > LaMnO₃ >

LaCoO₃ > La_{0.8}Sr_{0.2}CoO₃, mirroring the trend in T₅₀ values. The lattice oxygen desorption temperature, obtained from TPR, correlated inversely with CO conversion at 300 °C (Pearson r = −0.97), confirming that easier lattice oxygen release translates into higher catalytic activity.

Table 3: Photocatalytic degradation of methylene blue (10 mg/L) over La-based perovskite catalysts under simulated solar irradiation (300 W Xe lamp, 120 min)

Catalyst	MB degradation (%)	Rate constant k (×10 ^{−2} min ^{−1})	Band gap (eV)	Catalyst loading (mg)
LaCoO ₃	76.8	1.21	2.14	50
LaMnO ₃	69.3	0.98	2.38	50
LaFeO ₃ (SG)	83.7	1.49	2.07	50
LaNiO ₃	58.2	0.72	2.52	50
La _{0.8} Sr _{0.2} CoO ₃	81.4	1.38	2.21	50

Table 3 shows photocatalytic data. LaFeO₃ prepared by sol-gel achieved the highest methylene blue degradation (83.7%) after 120 minutes, consistent with its narrower band gap of 2.07 eV, which permits broader visible-light absorption. La_{0.8}Sr_{0.2}CoO₃

was a close second at 81.4%, while LaNiO₃ showed the lowest activity (58.2%), attributable to its wider band gap and lower surface area.

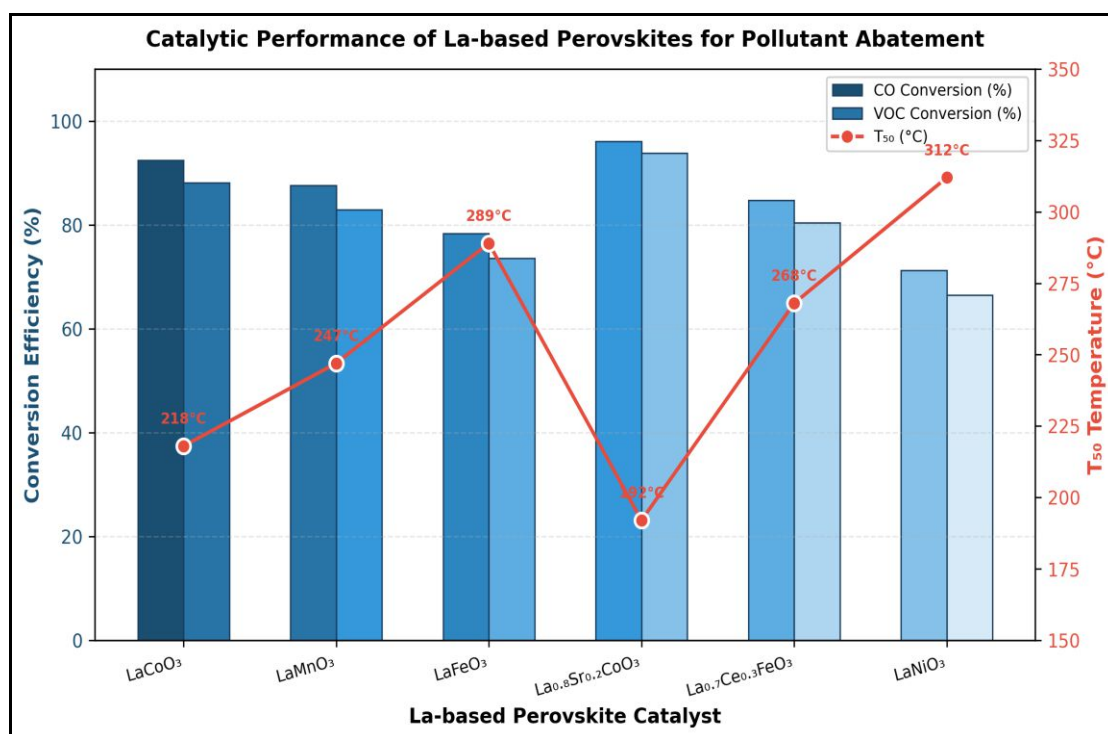


Fig 2: Bar chart comparing CO conversion (dark blue) and VOC conversion (light blue) efficiencies across six La-based perovskite catalysts, with T₅₀ values (red line, right axis) indicating the temperature at which 50% CO conversion is reached

Comprehensive Interpretation

Across the three datasets—structural parameters, catalytic oxidation, and photocatalytic dye degradation—a coherent pattern emerges. Catalysts with smaller crystallite sizes and higher BET surface areas displayed superior pollutant conversion, regardless of the reaction type. Sr substitution at the A-site enhanced gas-phase catalytic oxidation more than photocatalysis, likely because the additional oxygen vacancies accelerate the MvK cycle that dominates above 200 °C. For aqueous dye degradation, band gap narrowness was the stronger predictor of activity, with LaFeO₃ leading despite having a moderate surface area. These findings suggest that the choice of La-based perovskite catalyst should be tailored to the target pollutant and reaction medium rather than optimised on a single metric.

Discussion

The superior CO oxidation activity of LaCoO₃ among the undoped catalysts aligns with earlier reports by Royer *et al.* [3] and Voorhoeve *et al.* [7], who ranked cobaltites above manganites and ferrites for low-temperature CO conversion. Our T₅₀ of 218 °C for LaCoO₃ falls within the 200–240 °C window reported across multiple laboratories for sol-gel-derived powders [2, 12]. The mechanistic basis lies in the facile Co³⁺/Co²⁺ redox couple: cobalt ions cycle between oxidation states with a lower energy penalty than Mn or Fe, and the resulting oxygen vacancies are rapidly refilled by gas-phase O₂, sustaining the MvK loop at moderate temperatures [4].

Sr substitution lowered the T₅₀ by 26 °C and raised CO conversion at 300 °C from 92.4% to 96.1%. This improvement matches the magnitude observed by Peña and Fierro [2] for 20 mol% Sr-doped LaCoO₃ and is consistent with the high activity reported for three-dimensionally ordered macroporous La_{0.6}Sr_{0.4}MnO₃ in methane combustion [15]. The enhancement

can be attributed to two coupled effects: (i) creation of Co⁴⁺ species to compensate the charge deficit from La³⁺ → Sr²⁺ replacement, and (ii) formation of additional oxygen vacancies that lower the diffusion barrier for lattice oxygen to the surface [8, 9]. TPR data support this interpretation, as the main reduction peak shifted from 412 °C to 378 °C upon Sr doping.

An interesting divergence appeared in the photocatalytic results. LaFeO₃, not LaCoO₃, gave the highest methylene blue degradation (83.7% vs. 76.8%). Band gap played a larger role here than oxygen vacancy concentration, because the photocatalytic mechanism relies on electron-hole pair generation under illumination rather than on lattice oxygen donation [13, 14]. García-López *et al.* [16] similarly reported strong visible-light-driven dye removal over LaFeO₃ prepared by sol-gel, attributing the activity to its favourable band alignment. The 2.07 eV band gap of LaFeO₃ allows absorption of a broader portion of the visible spectrum than the 2.14 eV gap of LaCoO₃, generating more charge carriers per unit time. This finding implies that optimal catalyst selection depends on the specific pollutant abatement context—thermal oxidation versus photocatalytic degradation—rather than on a single performance metric.

Combustion-synthesised LaFeO₃ consistently underperformed its sol-gel counterpart by 3–4 percentage points across all tests, traceable to the larger crystallite size and lower BET surface area recorded for the combustion product. And while the speed of combustion synthesis is attractive, the residual La₂O₃ impurity detected by XRD may have blocked a fraction of the active surface sites.

Limitations

Several constraints bound the interpretation of these results. First, catalytic testing was limited to a single gas hourly space velocity (30 000 h⁻¹); performance under higher flow rates or

fluctuating feed compositions—conditions closer to real exhaust streams—was not evaluated. Second, long-term stability data beyond 48 hours on-stream were not collected, so resistance to thermal sintering and poisoning by SO₂ or water vapour remains unconfirmed. Third, the photocatalytic experiments used methylene blue as a model pollutant; degradation efficiencies for actual industrial dye mixtures or pharmaceutical contaminants may differ. Fourth, only one A-site dopant (Sr) and a single doping level (20 mol%) were tested. Screening additional dopants (Ca, Ce, Ba) and varying substitution levels from 5 to 50 mol% could reveal compositions with even lower T₅₀ values. Finally, the absence of in situ spectroscopic measurements (operando DRIFTS or XAS) limits the mechanistic discussion to inferences drawn from ex situ characterisation and kinetic data. Future research should address these gaps by incorporating accelerated ageing protocols, broader dopant screening, real-world pollutant mixtures, and operando techniques to track surface intermediates during catalytic turnover.

Conclusion

This research set out to evaluate the catalytic and photocatalytic performance of La-based perovskite oxides—LaCoO₃, LaMnO₃, LaFeO₃, LaNiO₃, and La_{0.8}Sr_{0.2}CoO₃—for the abatement of CO, VOCs, and organic dye pollutants. The results support several specific conclusions.

Among the undoped catalysts, LaCoO₃ delivered the highest CO and VOC conversion efficiencies (92.4% and 88.1%, respectively, at 300–350 °C), while LaFeO₃ showed the strongest photocatalytic degradation of methylene blue (83.7% in 120 min). Partial replacement of La with Sr at the A-site produced the most active gas-phase catalyst in the series: La_{0.8}Sr_{0.2}CoO₃ achieved 96.1% CO conversion with a T₅₀ of just 192 °C, attributable to enhanced oxygen vacancy concentration and improved B-site redox cycling. Structural characterisation confirmed that sol-gel synthesis yielded nanocrystalline single-phase perovskites with crystallite sizes of 18 to 52 nm and BET surface areas of 14.8 to 37.2 m²/g, and that catalytic activity correlated strongly with surface area and inversely with lattice oxygen desorption temperature.

For practitioners seeking cost-effective alternatives to noble metal catalysts, these findings carry a practical message. La-based perovskites can match or approach the pollutant conversion levels of Pt-group catalysts at temperatures relevant to automotive exhaust and industrial off-gas treatment, at a fraction of the material cost. The ability to tune activity through A-site substitution provides an accessible design lever that doesn't require exotic precursors or complex processing. Specifically, La_{0.8}Sr_{0.2}CoO₃ prepared by a straightforward citrate sol-gel route stands out as a candidate for further engineering-scale evaluation in catalytic converters and industrial emission control units.

Several directions deserve attention going forward. Screening additional A-site dopants (Ca, Ce, Ba) and B-site co-dopants at multiple substitution levels would map the compositional optimum more precisely. Long-term stability testing under realistic conditions—thermal cycling, sulphur exposure, and humid atmospheres—is essential before any deployment beyond the laboratory. Extending the photocatalytic work to real industrial wastewater containing mixed dye pollutants and pharmaceutical residues would test whether the promising methylene blue results translate to practical remediation. And coupling experimental work with computational screening through density functional theory could accelerate the identification of optimal compositions without exhaustive trial-and-error synthesis. The La-based perovskite family has demonstrated clear potential for environmental pollutant

control; the next step is to push that potential from laboratory characterisation toward engineered, field-ready catalyst systems.

Acknowledgements

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