

Perovskite Nanomaterial

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Recently, perovskite-based nanomaterials are utilized in diverse sustainable applications. Their unique structural characteristics allow researchers to explore functionalities towards diverse directions, such as solar cells, light emitting devices, transistors, sensors, etc. Many perovskite nanomaterial-based devices have been demonstrated with extraordinary sensing performance to various chemical and biological species in both solid and solution states. In particular, perovskite nanomaterials are capable of detecting small molecules such as O₂, NO₂, CO₂, etc. This review elaborates the sensing applications of those perovskite materials with diverse cations, dopants and composites. Moreover, the underlying mechanisms and electron transport properties, which are important for understanding those sensor performances, will be discussed. Their synthetic tactics, structural information, modifications and real time sensing applications are provided to promote such perovskite nanomaterials-based molecular designs. Lastly, we summarize the perspectives and provide feasible guidelines for future developing of novel perovskite nanostructure-based chemo- and biosensors with real time demonstration.

perovskite

nanomaterials

hybrid materials

chemosensory

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transistors

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real time application

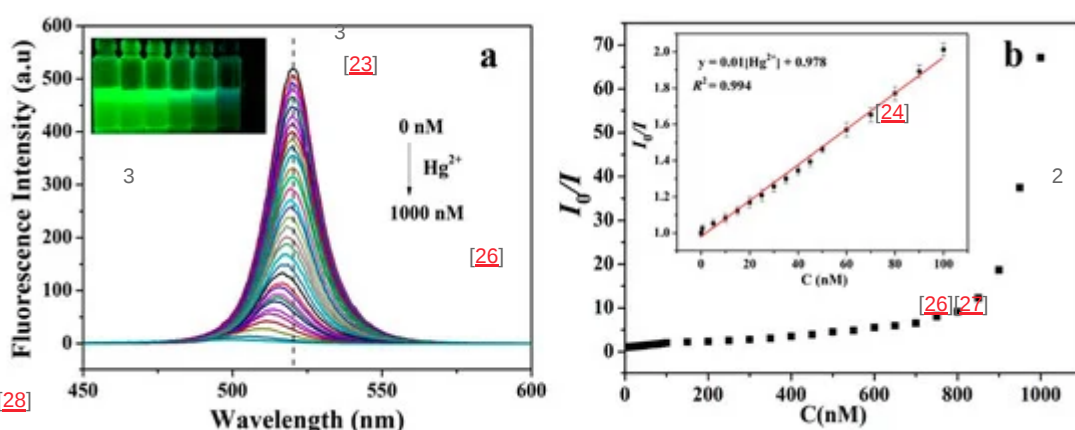
1. Definition

Perovskite is a kind of calcium titanium oxide mineral mainly composed of calcium titanate (CaTiO₃). Many different cations can be embedded in this structure, so a variety of engineering materials can be developed.

2. Introduction

Development of nanomaterials for diverse analyte detection with respect to environmental and biosafety measures are becoming essential ^{[1][2][3]}. Wherein, the species recognition can be identified by miscellaneous responses like colorimetric, spectrometry, voltammetry and morphological changes ^{[4][5][6][7][8]}. Among the reported nanomaterials, perovskites are exceptional hybrid materials with variety of applications, such as solar cells, light emitting devices, transistors, sensors, etc. ^{[9][10][11][12][13][14]}. The compounds that have the ABX₃ formula type with differently sized 'A' and 'B' cations bind to anion X are known as perovskite ^[15]. These perovskites are classified in three categories: inorganic oxide perovskites, alkaline metal halide perovskites and organic metal halide perovskites with oxide or halide anions ^{[16][17]}. Moreover, they can be synthesized from zero to three dimensional nanostructures and consumed in many sustainable applications ^{[18][19][20]}. Among these applications, sensory utilities using perovskite nanomaterials to attain the signals to specific analyte in solid or solution states have attracted most attention ^{[21][22]}.

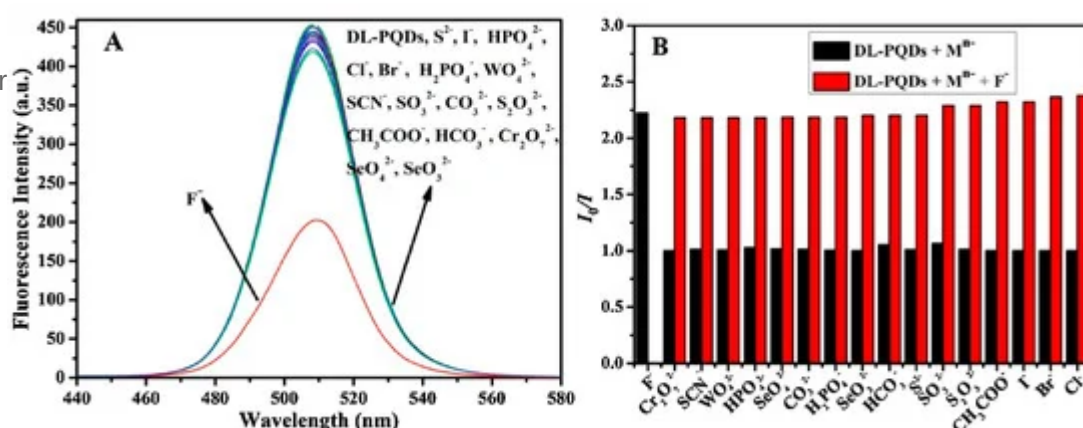
Perovskites consumed in the detection established the halide/hybrid fluorescence, advantage of metal halide temperature [28]



hence were perovskites allow Wang et al. as [25]. Metal fluorescence, also have the er, stability of ire, time and :tragonal and

orthorhombic may play a vital role in sensory studies [29]. Therefore, an in-depth discussion is required for the Figure 14 (a) shows the fluorescence spectra of CsPbBr₃ QDs upon the addition of different amounts of Hg²⁺. The concentration of Hg²⁺ from top to bottom: 0–1000 nM. (b) Linear fitting curve of I_0/I with Doping or mixing of other nanostructures/ions over perovskites tends to form a nanocomposite like structure [24], which can be further utilized as sensors [30]. For example, Cho and coworkers recently demonstrated the humidity sensing capability of the CsPbBr₃/BaTiO₃ nanocomposite [31]. Other than its potential sensory applications, dimensional property alteration [40], hence perovskite doped sensors also attracted attention from a research field. For instance, Lu et al. established the perovskite nanomaterials showed an exceptional humidity [32]. QDs synthesized from LAB procedure with PLQY demonstrated the improved CsPbBr₃ nanocrystals towards the sensing of overgapping by means of photoluminescence (PL) transfer [33]. This kind of photoluminescence (PL) transfer sensor design has been extensively been researched [34]. Many procedures have been explored to develop perovskite nanomaterials, including chemical synthesis, size ball-milling, combustion synthesis, sputtering, organo-metal halide reaction, etc. [34][35][36][37][38][39][40]. Moreover, it now becomes essential for finding applications of perovskite nanomaterials in sustainable research, such as solar cells, light emitting devices, transistors and sensors.

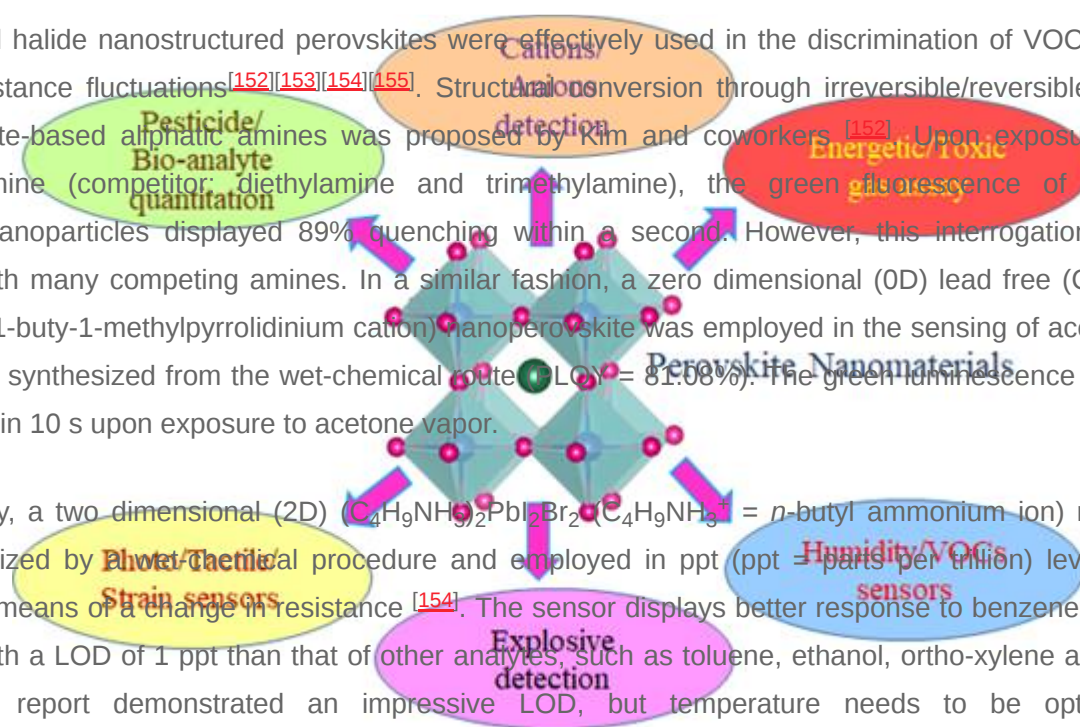
In this review, valuable information on sensory applications of perovskite nanomaterials (Figure 1) is provided. The mechanisms synthesis, structure and near future.



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Figure 15. (A) Fluorescence response of dual ligand capped perovskite quantum dots (DL-PQDs) to different anions and F⁻ (90 μM for F⁻ and 450 μM for other anions). (B) Interference studies of the novel nanosensor toward F⁻. The black bars represent the fluorescence response of DL-PQDs to F⁻ and other anions (90 μM for F⁻, 450 μM for other anions). The red bars represent the change of emission occurred after the subsequent addition of 90 μM of F⁻ to the above solutions (reproduced with the permission from reference [151]).

Organometal halide nanostructured perovskites were effectively used in the discrimination of VOCs by means of PL and resistance fluctuations^{[152][153][154][155]}. Structural conversion through irreversible/reversible H-bonding in the perovskite-based aliphatic amines was proposed by Kim and coworkers^[152]. Upon exposure to gaseous monoethylamine (competitor: diethylamine and trimethylamine), the green fluorescence of $\text{CH}_3\text{NH}_3\text{PbBr}_3$ perovskite nanoparticles displayed 89% quenching within a second. However, this interrogation needs to be upgraded with many competing amines. In a similar fashion, a zero dimensional (0D) lead free $(\text{C}_9\text{NH}_{20})_2\text{MnBr}_4$ ($\text{C}_9\text{NH}_{20}^+ = 1\text{-butyl-1-methylpyrrolidinium cation}$) nanoperoovskite was employed in the sensing of acetone^[153]. The material was synthesized from the wet-chemical route (LOD = 81.08%). The green luminescence was quenched (50 fold) within 10 s upon exposure to acetone vapor.



Subsequently, a two dimensional (2D) $(\text{C}_4\text{H}_9\text{NH}_3)_2\text{PbI}_2\text{Br}_2$ ($\text{C}_4\text{H}_9\text{NH}_3^+ = n\text{-butyl ammonium ion}$) nanoperoovskite was synthesized by a wet-chemical procedure and employed in ppt (ppt = parts per trillion) level detection of benzene by means of a change in resistance^[154]. The sensor displays better response to benzene ($R_g/R_a = 90.7$) at 160 °C with a LOD of 1 ppt than that of other analytes, such as toluene, ethanol, ortho-xylene and para-xylene gases. The report demonstrated an impressive LOD, but temperature needs to be optimized before commercialization. In this framework, the methylammonium lead iodide (MAPbI_3) nanostructured perovskite thin film has been proposed to sense ethanol gas at room temperature^[155]. Both the resistance and PL responses can be used for the determination of ethanol. However, the LOD of this ethanol sensor is approximately 1300 ppm, which require further improvement in perovskite-based sensing of VOCs.

Figure 1 Schematic illustration of the sensory applications of perovskite nanomaterials

3. Structure, Stability and Properties of Perovskites

Similar to the VOCs sensing, perovskites can be an excellent candidate in gas discrimination. Methylammonium lead tri-iodide (MAPbI_3) hybrid perovskites display its high sensitivity to ammonia (NH_3) gas^{[156][157][158]}. Perovskites with ABX_3 formula exist in a undistorted octahedron structure with MA⁺ to coordinate of small separation provided by a **octahedron of anions** stretching down the 12 fold octahedral coordinated ability of MA⁺ cations was established. Figure 2. Metal oxide perovskites exist of $\text{MA}(\text{Pb}, \text{Sn})\text{X}_3$ dimensional frameworks with $n=1$ and 5.5 with high response values (R_g/R_a of 20 to 500 ppm)^[159]. Instead of low gas synthesized metal halide perovskites, A-site the displayed the defective distorted by tilting and 200 pm the octahedron by cation displacement, which leads to moderate symmetries like **orthorhombic**, **tetragonal** and **trigonal**. In the framework of the perovskite structure, the base with the cubic structure, but they also require MA⁺ to coordinate tolerance factor with values between 0.8 and 1.0. MA⁺ to coordinate factor is played an important role in the response to NH_3 gas, as illustrated in **Figure 1.6** and NH_3 presented the respective A, B and X sites^[41]. If the tolerance factors of the perovskite materials have a value between 0.7 and 0.9, they could have a distorted substructure, halide perovskites are of symmetries. Effective in the also, factors the stability of many metal halide perovskites is also affected by other factors, such as temperature, water and the environment as described subsequently.

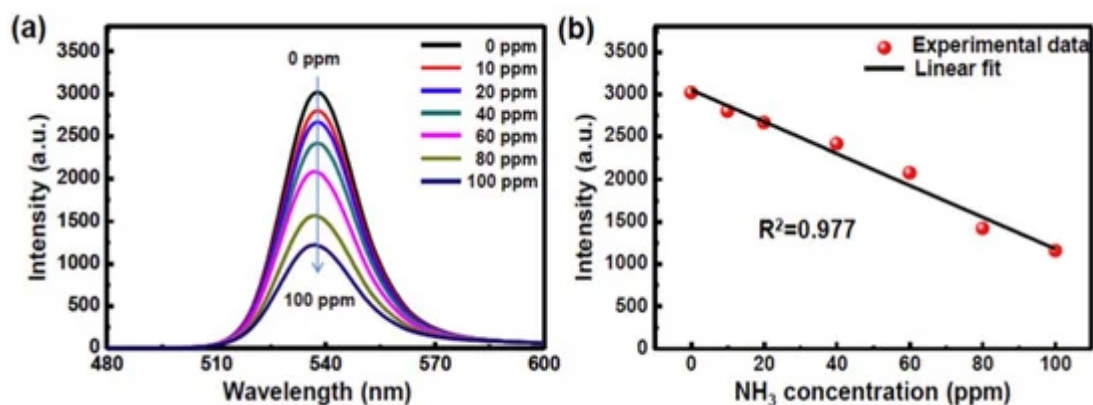


Figure 16. MAPbBr₃-TBA-based gas sensor. (a) The PL quenching toward different concentration of gaseous NH₃ (0–100 ppm) and (b) the plot of PL intensity versus concentration of NH₃ (reproduced with the permission from reference [160]).

Since annealing is an important step in the fabrication processes of metal halide perovskites for sustainable applications, it is generally halide perovskites were also utilized in the device-based assay of toxic NO₂ gas, the majority of metal halide perovskites might experience phase transitions. For example, CH₃NH₃PbBr₃ and CH₃NH₃SnBr₃ showed high selectivity to NO₂ with LODs of 200 ppb, 11 ppm and 1 ppm and 25 ppb respectively, via resistance changes. [163][164][165][166] Moreover, CH₃NH₃Pb(SCN)₃ also displayed a sensing ability to gaseous acetone with a LOD of 20 ppm. The underlying mechanism is that the NO₂ gas is physically adsorbed on the surface of CH₃NH₃PbBr₃ (those films generate electrons from the conduction band) or from charge transfer chemisorption. A trihalide ion containing FA⁺, MA⁺, Cs⁺, Pb²⁺, [45][46][47][48][49] CH₃PbBr₃ was a crystalline perovskite was applied in the device-based detection of NO₂ gas by Chen and coworkers. [166] As shown in Figure 17, FAPbBr₃ devices in the self- and externally-powered mode were able to detect NO₂ gas at room temperature with an estimated LOD of 0.2 ppm. This material is a The stability of perovskites is mostly affected by water, which cause the dissolution/degradation of materials during the fabrication process. In the case of metal oxide perovskites, they tend to form hydroxyl ions (OH⁻) over their surface with water, which is currently applied in the water splitting application [50]. On the other hand, in the presence of water, the metal halide perovskites may degrade due to the distortion of their lattice sites. Likewise, organic–inorganic hybrid metal halide perovskites were also affected by the existence of water molecules. For example, the CH₃NH₃PbI₃ decomposed into CH₃NH₃I and PbI₂ when encountered with the water molecules [51]. However, such degradation was also extensive in supportive environment.

Moisture environment or organic solvents in their gaseous state also significantly affect the stability of perovskite materials [52][53]. The stability of perovskite materials was considerably disturbed when exposed in a gaseous environment, such as NO₂, CH₄, NH₃, C₂H₅OH, acetone, etc. [26], as a result, they can be used as sensors for those gaseous species. The abrupt changes in these perovskite materials can be recorded through chemiresistive I–V, phosphorescence and fluorescence responses. Nevertheless, the opto-electronic properties of perovskites play a vital role in these sensing studies.

Perovskite oxides are well known candidates with exceptional properties, such as electrical conductivity, ferroelectricity, superconductivity, catalytic activity, etc. For example, the studies on the ferroelectricity of BaTiO₃

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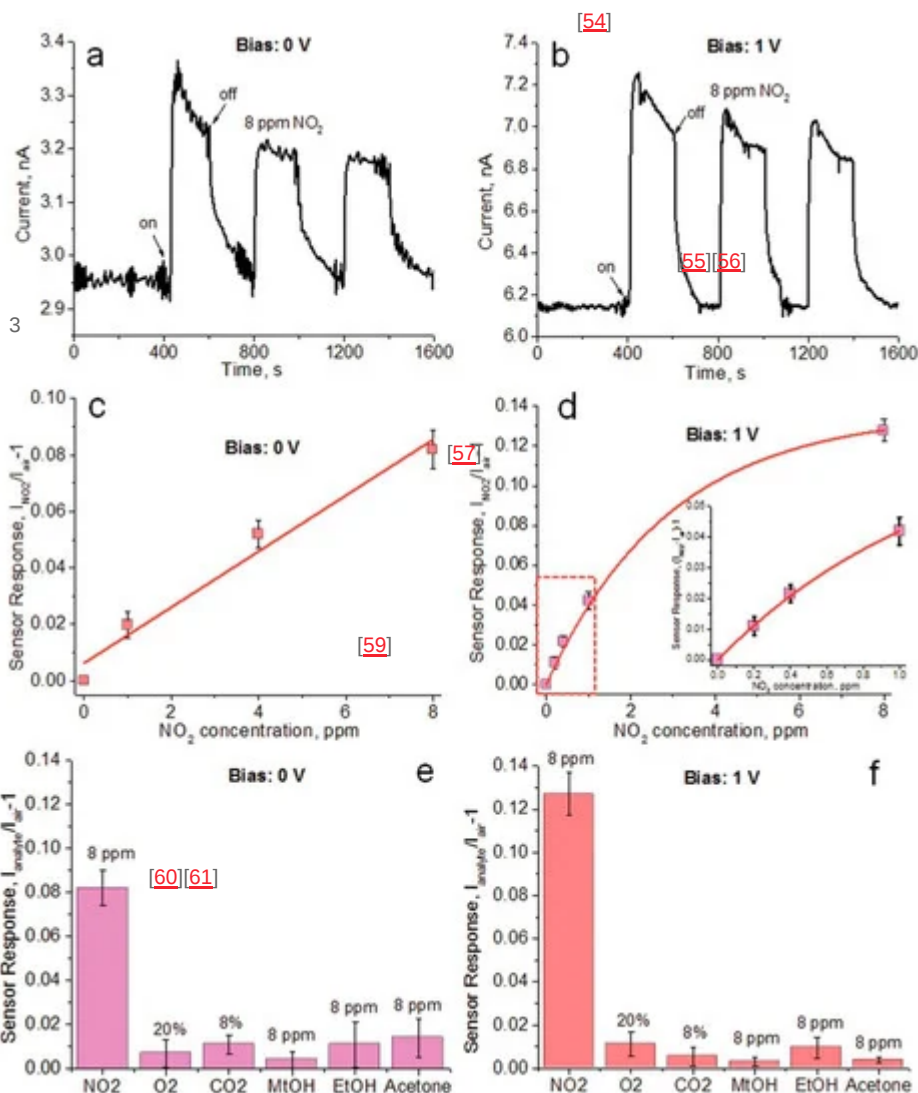
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expansion. Similarly, ferroelectric properties of halide perovskites have been investigated in many reports [63]. Enhancement in photovoltaic properties by means of ferroelectric domain structures in MAPbI₃ was demonstrated by Yamada and coworkers [64]. Response of an FMCPB device in response to three consecutive injections of 8 ppm NO₂ (a) [54] have directed researchers towards optical sensor investigations [65] and (b), externally powered with the V bias in the dark, FMCPB sensor response as a function of the NO₂ concentration [66]; (c) in the self-powered mode and (d) at the 1 V external bias; the inset in (d) is the enlarged part of the red dashed region. The sensor response of the FMCPB sensor for six different gases (e) in the self-powered operational mode; and (f) at the 1 V external bias. The gas sensing measurements were conducted at room temperature (30 °C) under simulated air (V_{N2}/V_{O2} = 4) with a constant total gas flow of 0.5 L min⁻¹ (reproduced with the permission from reference [166]).

4. Factors Affecting Sensor Interrogations of Perovskite Nanomaterials

Sensory utilities of perovskite nanomaterials can be affected by the following factors, hence the design of suitable Organometal halide nanoperoovskites were engaged in recognition of O₂, O₃ and H₂ gases, as well as photo nanostructures towards a specific direction must be done with the consideration of these elements. sensors [167][168][169][170] through conductivity responses. For example, Stoeckel et al. used the CH₃NH₃PbI₃

nanocrystalline film to determine O₃ concentration via the trap healing mechanism instigating from an O₂-tuned iodine vacancies filling with a detection limit down to 70 ppm [167]. This is an inspiring work on device-based O₃ still a question to researchers. Since perovskites may form diverse nanostructures such as quantum dots, nanocrystals, nanowires/rods, nanoplates, etc. It is still the most difficult challenge for scholars to identify the proper perovskite nanomaterials for their target sensor investigation. Another critical issue is that some synthetic responses of this sensor material in the presence of both gases (O₃ and H₂) need to be established for validation.



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Next, the $\text{CH}_3\text{NH}_3\text{PbI}_3$ thin film of crystalline structure was grown by the above strategy deposited the (PbI₂) process, which afforded explicit nanostructured materials with lengths from 375 to 800 nm at 5 V bias [170]. This material also exhibited a power conversion efficiency (PCE) of 10.6% and became a potential candidate in photonics research.

Stability: perovskite nanomaterials has the major issue of stability, which might influence many sensor responses.

For example, the sensing ability of halide perovskites have been significantly affected by moisture and toxic gases and exposure to the oxide and sulfide perovskite chemical sensors. unstable by temperature, Xue et al. reported the two-dimensional (2D) $(\text{CH}_3\text{NH}_3)_2\text{PbI}_4$ based strain sensor, which displayed high sensitivity and a strain ratio of 0.316% with reversible stretchability [68][69][79]. These factors that hinder the breakthrough in structural sensor technology.

By sensing design, some materials that require nanoprecursors. Dual halide perovskites may form the composite perovskite structures, which still can be utilized in electronic devices based like sensors [172][173].

For instance, the nanocomposite like structure with a combination of ZnO nanosheet arrays and polystyrene (or $\text{CH}_3\text{NH}_3\text{PbI}_3$ (MAPbI₃)) acts as exceptional tactile sensors (with respective sensitivity of 0.57 kPa⁻¹ and 0.64 kPa⁻¹). The sensing applications of nanocomposites with perovskites are discussed in the next section.

Toxicity or environmental affordability: Toxicity measurements may tell us the biocompatibility of those materials to be consumed in healthcare products. However, majority of halide perovskites

are likely to be toxic, hence their use in biosamples are rather restricted. For example, $\text{CH}_3\text{NH}_3\text{PbX}_3$ (X = Cl, Br and I) are well known candidates with good emissive nature but should be avoided to use in biosamples.

Bio/environmental samples may be affected by the presence of toxic Pb ions, hence actions are needed to Metal oxide perovskites incorporated nanocomposites displayed high sensitivity to ethanol gas with exceptional eliminate their harmfulness via suitable modifications with appropriate capping or cations [74].

response and recovery [92 via suitable]. Wherein, $\text{LaMnO}_3/\text{SnO}_2$ composite nanofibers [174] and $\alpha\text{-Fe}_2\text{O}_3/\text{LaFeO}_3$ composite nanomaterial [175] delivered their sensitivity to 100 ppm ethanol at high operating temperatures 260 and 240 °C with responses of 20 and 10, respectively. In contrast, Chen et al. reported the $\text{Ag}/\text{Zn-LaNiO}_3$ (AZLFO) modern research topic. However, developing such luminescent materials with analyte specificity is still a challenge, nanocomposite-based ethanol detection, which showed a response of 64.2–100 ppm analyte and a detection limit Since luminescent property may vary at diverse precursor dilution [72], it is very essential to develop materials with down to 5 ppm [176]. Note that this sensor can operate from 55 to 245 °C. By following the reaction steps shown in high quantum yield (Φ) values. For example, Zhu et al. publicized the CsPbBr_3 perovskite nanocrystals with 87% Equations (3)–(5), ethanol sensing can be achieved near room temperature.

quantum yield towards colorimetric sensing of peroxide number in edible oils [73]. Therefore, the development of

luminescent perovskite nanomaterials with high quantum yield is expected for sensor studies.



5. Sensing Utilities of Metal Oxide Perovskite Nanomaterials



Until now, many semiconducting perovskites have been reported in a variety of gas sensing studies, which can be applied in environmental, fire and vehicle monitoring [74]. This might be attributed to the interaction of analyte gases. It has been established that the gas sensitivity is achieved due to fluctuations in electrical conductivity induced by oxygen presented in the perovskite grain boundaries, which results in a fluctuation in electrical conductivity [75]. surface chemisorbed oxygen species over the sensing materials. However, the Zn doping also plays a vital role for in this path, the majority of metal oxide perovskites were utilized towards the detection of various gaseous or near room temperature sensing of ethanol. As illustrated in Figure 18, Zn^{2+} doping compensates the defects of La hazardous volatile species [76][77][78][79][80][81][82][83][84][85]. In addition, numerous reviews and book chapters have sites and increases the hole concentration, thus a higher sensor response is achieved near room temperature, explored and demonstrated these sensing applications in detail [86][87][88][89][90]. Therefore, the recently published sensory studies were mostly focused in this field.

Sensing responses of metal oxide perovskites are majorly attributed to the doping of ions or the composite mixture.

Cao and coworkers reported the chlorine-doped nanocrystalline LaFeO_3 powders towards ethanol gas sensing via resistance change [91]. They employed the citric sol–gel method to vary the chlorine doping in LaFeO_3 , which enhanced the sensing performance via improved grain size and reduced intrinsic resistance. At 136 °C, LaFeO_3 .

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nanoplates was estimated as 50 ppm (ppm parts per million) in a wide range of humidity from 20 to 90%. This work

was impressive, but the operation temperature still required to be reduced. Recently, Cao and coworkers proposed **Figure 18**. Schematic illustration of the sensing mechanism of the AZLFO-based sensors to ethanol: (a) exposed using the Au and Cl co-modified LaFeO_3 nanoparticles (size = 29.5 nanometer (nm)) for the detection of ethanol to ethanol; (b) exposed to air and (c) Zn^{2+} doping in the lattice of LFO (reproduced with the permission from reference [176]).

method and the sensor signal was attained via resistance change. They improved the ethanol sensing

characteristics of Cl-doped LaFeO_3 [91] by the inclusion of the Au atom.

Such a composite structure has been applied in the sensing of gaseous acetone [177]. Three dimensional (3D)

$\text{LaFeO}_3/\alpha\text{-Fe}_2\text{O}_3$ nano-octahedrons were synthesized by the one-step solvothermal method, which were then Growth of miscellaneous nanostructured metal oxide semiconducting perovskites to detect assorted gaseous combined with a metal-organic framework. The nano-octahedrons constructed heterostructures detect the acetone species has recently attracted much attention. For instance, MA et al. synthesized the *p*-type PrFeO_3 and obtain the sensor response ($R_a/R_0 = 21$) by means of changes in conductivity. The sensor can detect 100 ppm (praseodymium ferrite) mesoporous hollow nanofibers through electrospinning and calcination procedures and of acetone at 230°C , thereby further optimization is required to reduce the working temperature. Porous SnO_2 employed in gaseous acetone discrimination [96]. When exposed to 200 ppm of various gases at 180°C , PrFeO_3 fiber-in-tubes (FITs) were functionalized with $\text{La}_{0.75}\text{Sr}_{0.25}\text{Cr}_{0.5}\text{Mn}_{0.5}\text{O}_{3-\delta}$ (LSCM) nanoparticles (215.7 nm in size; nanofibers showed exceptional selectivity to acetone ($R_g/R_a = 141.3$) with long term stability as shown in Figure 3. synthesized from the combustion method using citric acid) and employed in formaldehyde recognition [178]. The Oxygen in air was adsorbed on the surface of PrFeO_3 to capture the electrons of materials and increased the hole material (LSCM@ SnO_2 FITs) showed a high response to formaldehyde ($R_a/R_0 = 26.50$ at 5 ppm, 400°C) with a concentration, hence the resistance decreased. However, when the acetone gas entered, it interacted with LOD of 80 ppb. The above report is impressive work but the operation temperature must be reduced for practical chemisorbed oxygen and released the electrons to recombine with holes, which resulted in increased resistance applications. Later, hydrothermally synthesized nanoflowers like the $\text{ZnSnO}_3/\text{Zn}_2\text{SnO}_4$ composite hybrid has been (this mechanism is applicable to the majority of volatile organic compounds (VOCs)). Moreover, PrFeO_3 hollow nanofibers also displayed linear resistance change from 10 to 500 ppm acetone gas. Therefore, one can certainly of 12.1–20 ppm phenylamine (at 260°C) with a LOD of 50 ppb and response/recovery time of 1 s/20 s. Due to its endorse the potential acetone sensing ability of PrFeO_3 hollow nanofibers. anti-humid property, these materials can be employed in the determination of toxic phenylamine.

Investigation on SmFeO_3 -modified MoS_2 ($\text{SmFeO}_3@\text{MoS}_2$) nanocomposites towards humidity sensing has been conducted by the Zhang research group [180]. The material, synthesized by electrospinning combined with the hydrothermal technique, operates from 11 to 95% RH with a recovery/response time of 1.5 s/29.8 s, thus it can be considered as an effective candidate. Other than the VOCs and humidity sensors, perovskite enabled composites were utilized in toxic gas quantitation. For example, $\text{La}_{0.8}\text{Sr}_{0.2}\text{FeO}_3$ (LSFO) nanoparticles (with a size of 100–300 nm) decorated Ga_2O_3 nanorod arrays have been employed in the recognition of carbon monoxide (CO) via

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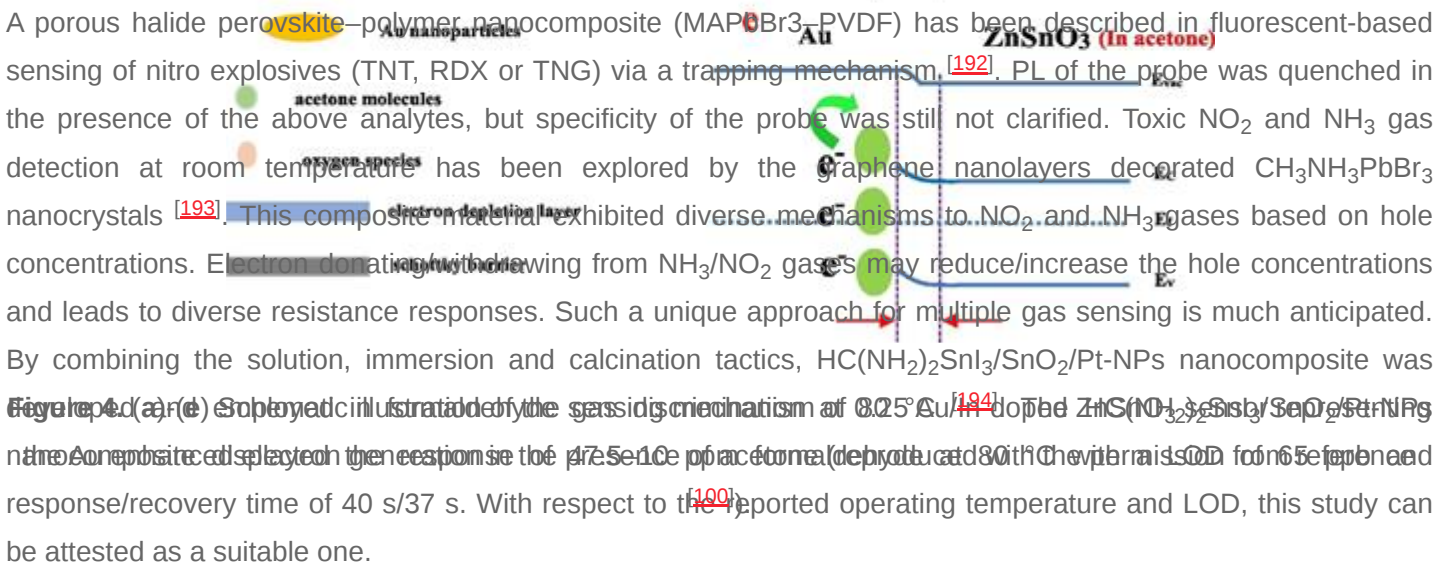
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conductivity studies [181]. The sensor operates at 500 °C with a response comparable to the Pt nanoparticles decorated Ga₂O₃ nanorod arrays. However, investigations on competing gases still need to be performed with this material. Joshi et al. reported work on BaTiO₃ included nanocomposites (Ag@CuO/BaTiO₃, CaO-BaTiO₃ and CuO/BaTiO₃) towards CO₂ gas sensing [182][183][184]. In fact, these materials used in the discovery of CO₂ gas showed an optimum sensitivity of 700, 1000 and 5000 ppm, correspondingly, at either 120 °C or 160 °C with a good response and recovery time, thereby confirmed the success of BaTiO₃ enabled composites for CO₂ recognition.

Toxic SO₂ gas detection has been established by wet chemically synthesized SnO₂/LaFeO_{3-x}N_x nanocomposite materials [185]. However, the response and other details on LODs and sensitivity were not clearly specified in this report. Recently, La_{0.8}Sr_{0.2}CoO₃ (LSCO) nanoparticles decorated β-Ga₂O₃ nanorod arrays were consumed in the detection of toxic SO₂ gas at 800 °C by Zhang et al. [186]. This work with desirable performance in sensitivity (6625 ppm) at high temperature 180 °C seems to be linear from 21.62 to 100 ppm with a LOD of 1 ppm. A 3.38 times higher response than pristine ZnO was achieved using Ag functionalized ZnSnO₃ nanocubes in acetone gas sensing. This work is an impressive one in terms of the detection limits, but the operating temperature still requires further optimization. Through the sol-gel method, researchers fabricated the Pd-doped SnFe_{1-x}Mg_xO₃ (x = 0, 0.1, 0.2 and 0.3) nanocrystalline powder and La_{1-x}Y_xMnO₃ (x = 0 and 0.15) nanoparticles towards acetone gas detection [98][99]. Even though both materials demonstrated excellent responses, the operating temperatures (220 °C and 300 °C) still need to be reduced. To this direction, Ag functionalized indium-doped ZnSnO₃ nanofibers were fabricated via electrospinning technique [100], which displayed sensitivity to 50 ppm acetone at 200 °C with a fast response/recovery time (10 s/13 s). As shown in Figure 3, the organic metal halide perovskites were also employed in many composite-based sensory applications.

Chemically synthesized metal ligand stabilized CdTeS₂ QDs (CPBQDs, PLQY = 28%) encapsulated in polyvinyl methacrylate (PMMA) nanofiber membrane (CPBQD/PMMA FM) via the electrospinning method were used in the sensing of trypsin, Cu²⁺ and pH [190]. As illustrated in Figure 19, the CPBQD/PMMA FM detects trypsin through the cleavage of peptide CF6 (Cys-Pro-Arg-Gly-R6G) followed by a Fluorescence Resonance Energy Transfer (FRET) between the fiber and cyclam-Cu²⁺, which leads to Cu²⁺ recognition. Finally, 10 ppb hydrazide R6G (in ethanol) plays a vital role in pH sensors. LODs of trypsin of 0.1 μg mL⁻¹ and Cu²⁺ quantitation of 10⁻¹⁵ M were reported. CH₃NH₃PbBr₃ QDs were incorporated in metal-organic framework (MOF-5) microcrystals and applied in temperature and heavy metal ions detection [191]. The CH₃NH₃PbBr₃@MOF-5 composites possess a wide range of pH adaptability and display its sensitivity to temperature from 30 to 230 °C. However, this probe showed sensor responses to many metal ions, thereby more work is required to achieved specificity.

Figure 3 (a) The response of ZnSnO₃ to 50 ppm acetone at 180 °C and (b) the work with desirable performance in sensitivity (6625 ppm) at high temperature 180 °C. Similar to the work with desirable performance in sensitivity (6625 ppm) at high temperature 180 °C, composites with perovskites were also employed in electrochemical sensors. Sr₂PdO₃ nanoperovskite mixed with carbon nanotubes (CNTs) were casted over a glassy carbon (GC) electrode surface (GC/CNTs/Sr₂PdO₃) and used in the electrochemical determination of dobutamine (DB), cardiac stimulator drug [137]. This sensor showed dynamic response to 100 ppm with a LOD of 1 ppm. A 3.38 times higher response than pristine ZnO was achieved using Ag functionalized ZnSnO₃ nanocubes in acetone gas sensing. This work is an impressive one in terms of the detection limits, but the operating temperature still requires further optimization. Through the sol-gel method, researchers fabricated the Pd-doped SnFe_{1-x}Mg_xO₃ (x = 0, 0.1, 0.2 and 0.3) nanocrystalline powder and La_{1-x}Y_xMnO₃ (x = 0 and 0.15) nanoparticles towards acetone gas detection [98][99]. Even though both materials demonstrated excellent responses, the operating temperatures (220 °C and 300 °C) still need to be reduced. To this direction, Ag functionalized indium-doped ZnSnO₃ nanofibers were fabricated via electrospinning technique [100], which displayed sensitivity to 50 ppm acetone at 200 °C with a fast response/recovery time (10 s/13 s). As shown in Figure 4, the organic metal halide perovskites were also employed in many composite-based sensory applications.



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ppb ($R_g/R_a = 1.7$). The above material can be consumed towards the discrimination of *n*-propanol in the presence of other analytes, such as acetone, xylene, ammonia, methane and hydrogen.

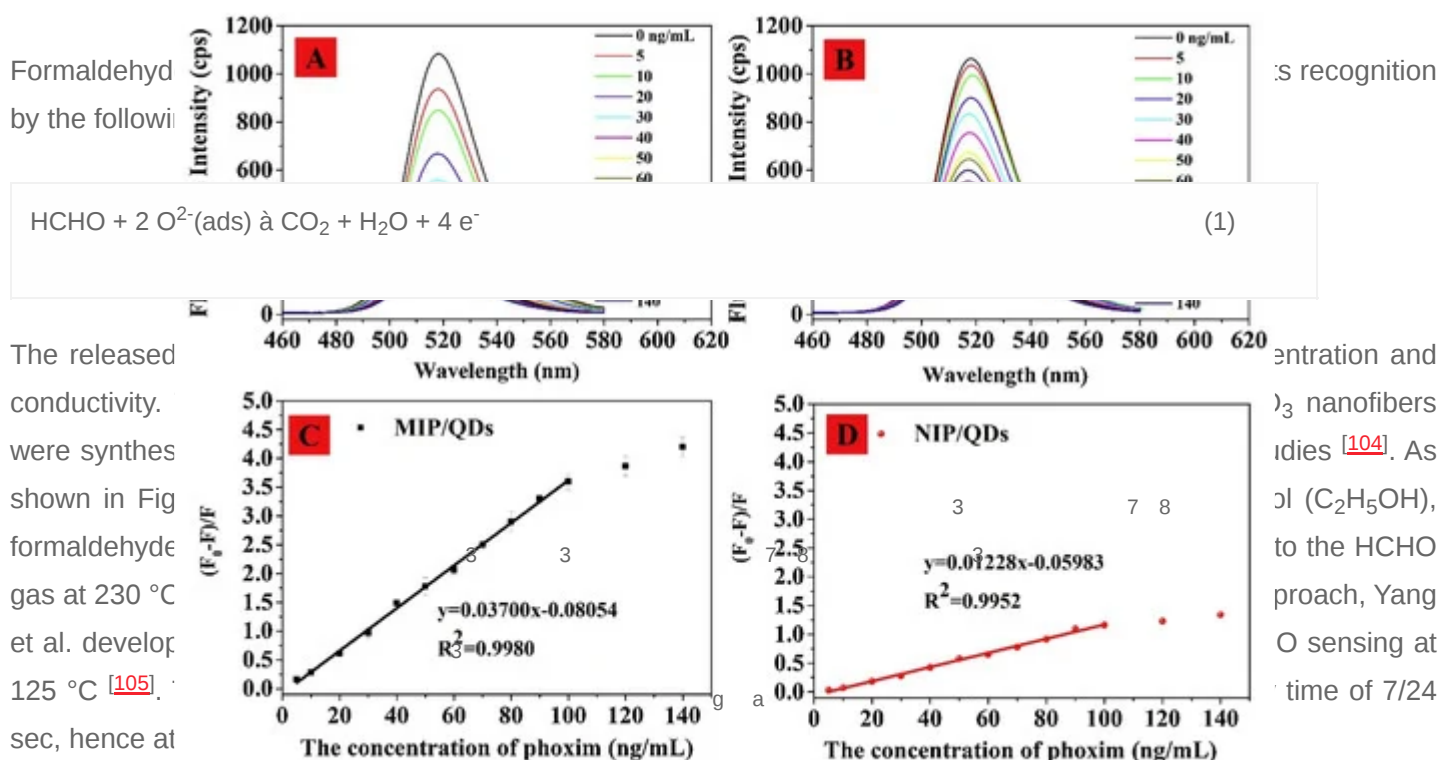


Figure 20. (A,B) Effect of phoxim concentration (0–140 ng/mL) on the fluorescence spectra of molecularly imprinted polymer MIP/QDs and NIP/QDs (25 °C). (C,D) Stern–Volmer plots of MIP/QDs and NIP/QDs with phoxim (reproduced with the permission from reference [196]).

In this light, CsPbBr₃/PQD immobilized TiO₂ inverse opal photonic crystals³(IOPCs³) have been engaged as electrodes for the electrochemical dopamine discovery [197]. This composite electrode expresses a linear response to dopamine from 0.1 to 250 μM with a LOD of 0.012 μM. The underlying mechanism of this sensor is discovered as “photonic stop band effect” of TiO₂ IOPCs on the incident light and the emission of PQDs, which enhances the photocurrent upon exposure to dopamine. This work allows the consumption of PQDs in a bioanalysis. In a similar fashion, nanostructured quasi-2D and 3D CH₃NH₃PbI₃ enabled TiO₂ film and molecularly imprinted polymers (MIPs) and polyethylene glycol (PEG) coated CH₃NH₃PbI₃ (MIP-PEG/CH₃NH₃PbI₃) were consumed in the electrochemical determination of carbon tetrabromide (CBr₄) and salicylic acid, correspondingly [198][199]. The quasi-2D and 3D CH₃NH₃PbI₃/TiO₂ composites can detect the CBr₄ down to 20 ppb mol⁻¹, and hence become a reliable system for CBr₄ sensing. Subsequently, the MIP-PEG/CH₃NH₃PbI₃ nanocomposite displayed remarkable sensing performance with high sensitivity to SA in the dynamic range of 1.0 × 10⁻¹⁰ to 1.0 × 10⁻⁸ mol/L and LOD and the limit of quantitation (LOQ) of 2.71 × 10⁻¹⁰ mol/L and 9.02 × 10⁻¹⁰ mol/L, respectively.

nanofibers doped by different amounts of Ag were marked as NF0, NF2, NF4 and NF6 (reproduced with the permission from reference [104]).

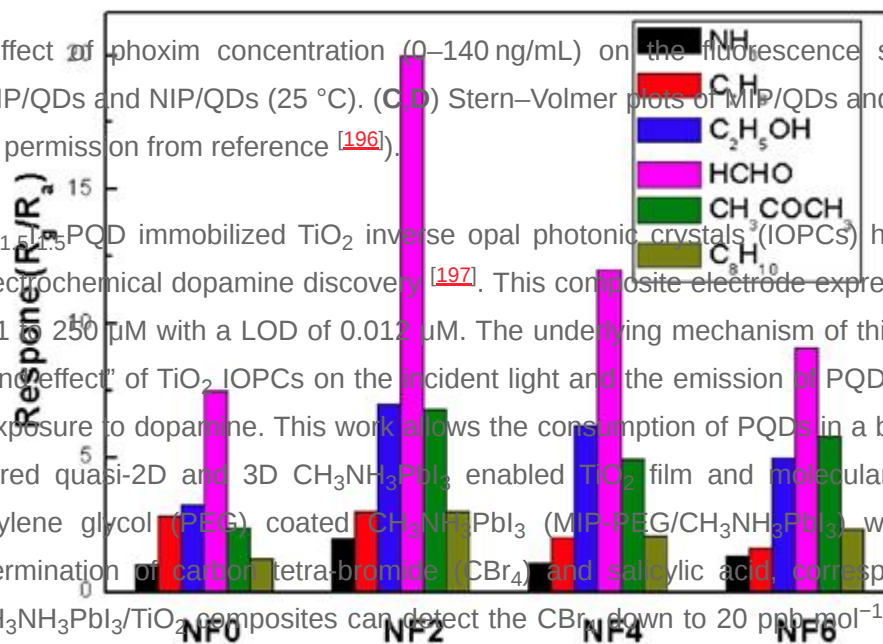
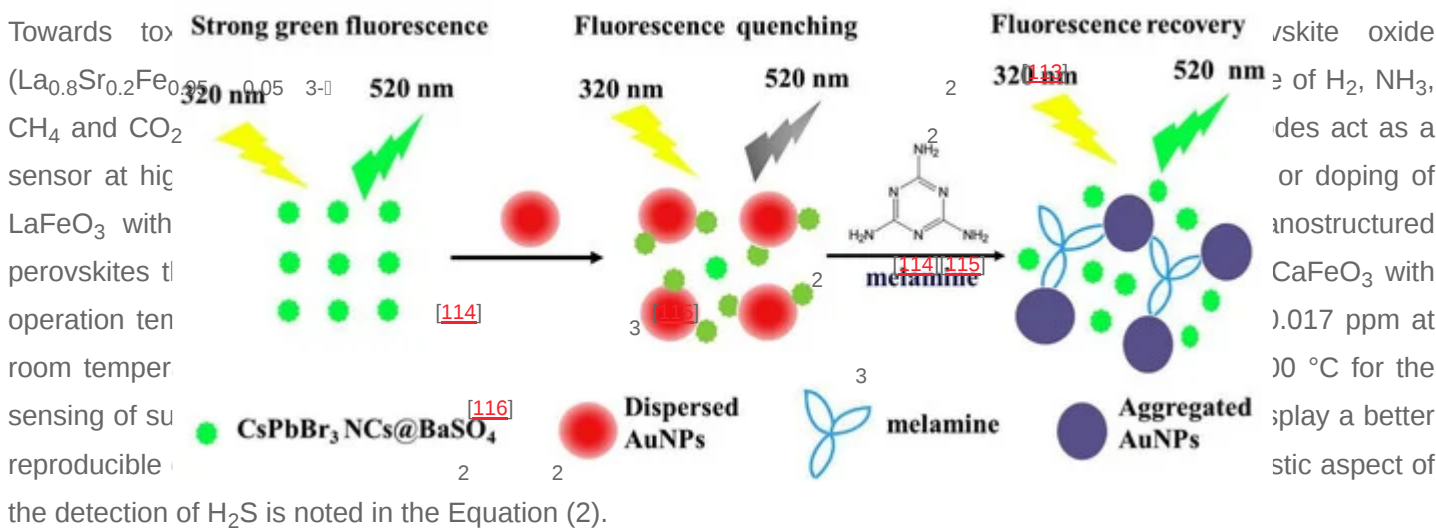


Figure 5. Responses of the sensors to 100 ppm different gases at the optimum working temperature. NF0, NF2, NF4 and NF6 (reproduced with the permission from reference [104]).

Recently, an inner filter effect-based melamine assay has been demonstrated by barium sulfate-coated CsPbBr₃. On the other hand, flameable toxic gas detection was also explored by many metal perovskite nanostructures (CsPbBr₃/Cs₂Se/BaSO₄) and AuNPs [200]. As shown in Figure 21, in the presence of AuNPs, the PL intensity of CsPbBr₃/Cs₂Se/BaSO₄ was quenched and then recovered upon addition of melamine. This was proposed by Bal and coworkers [106]. At 15% Au doping, BiFeO₃ showed a good sensor response (212% for 500 ppm) displayed the linearity from 5 to 500 ppmol/L with a LOD of 3.42 nmol/L. Moreover, this work also completed

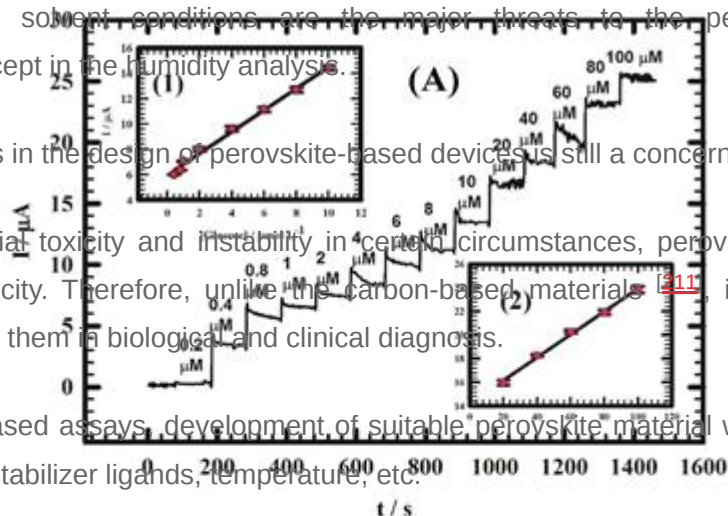

$$2\text{H}_2\text{S} + 3\text{O}_2 \rightarrow 2\text{SO}_2 + 2\text{H}_2\text{O} + 3\text{e}^- \quad (2)$$

With the optimum LOD (4 ppm) and response/recovery time (60–360 s/180–500 s), LaFeO₃ nanofibers are the best designed to be employed in the superkites-based gas sensors. Superkites-based gas sensors have certain advantages and limitations as listed below.

The non-enzymatic/enzymatic determination or direct recognition of analytes, such as glucose, *p*-phenylenediamine and H₂O₂ has been demonstrated using modified metal oxide perovskite electrodes [120,121,122]. A luminescent characteristic of metal halide/organometallic halide quantum dots or nanocrystals [123,124] advantages over Pt-based identification of specific target. Their metallic and crystalline nature further improve their sensitivity in comparison with carbon dots [210]. Cu⁺, Au³⁺ and Pt²⁺, respectively [119][121]. As shown in

Figure 6. Selectivity and sensitivity of both graphite/Sr₂Pd_{0.7}Au_{0.3}O₃ and Sr₂Pd_{0.7}Au_{0.3}O₃ (x = 0–0.7) with LOD of 0.202 μ M (inset with) and 2.1 μ M (main plot), successively. This work can be used in the electrochemical catalytic estimation of glucose. Toxic *p*-phenylenediamine (PPD) in hair dyes was electrochemically detected by Sr-doped PbCoO_3 ($\text{Pb}_{1-x}\text{Sr}_x\text{CoO}_{3-\delta}$ ($x = 0, 0.2, 0.4, 0.6, 0.8$ and 1)) modified glassy carbon electrodes (PSC82/GCE) in alkaline solution [120]. The electrode displayed highest responses of 655 and 308 $\mu\text{A mm}^{-2}$ in the PPD concentration range of 0.5–2.9 mM (millimole) and 2.9–10.4 mM, individually, with a LOD of 0.17 μM . However, this work still needs more effort to optimize the range of detection and LOD. Similarly, Y-doped (8% mol) SrTiO_3 displayed electrochemical sensitivity to H_2O_2 with a LOD of 14.97 μM [121], which also requires further improvement.

- Environment and solvent conditions are the major threats to the perovskite-facilitated sensory investigations, except in the humidity analysis.
- Cost-effectiveness in the design of perovskite-based devices is still a concern for the researchers.
- Due to the material toxicity and instability in certain circumstances, perovskite sensors have limits in reliability and toxicity. Therefore, unlike the carbon-based materials [211], it still remains an important challenge to apply them in biological and clinical diagnosis.
- For fluorescent-based assays, development of suitable perovskite material with high PLQY is limited by synthetic tactics, stabilizer ligands, temperature, etc.



9. Conclusions and Perspectives

This review summarized the sensing applications of metal oxide and metal halide organometallic halide perovskite nanomaterials. We focused on detection of toxic/energetic gases, humidity, VOCs and electromechanical quantitation using the perovskite nanomaterials-based devices and explained the underlying mechanisms. The fluorescent-based assays of analytes were also discussed in detail with given advantages and limitations. Moreover, the sensory utilities of perovskite-enabled nanocomposites were also reviewed. Besides the aforementioned sensing applications, the following points are yet to be focused.

- The underlying mechanisms in many sensory reports still require in-depth investigations with respect to theoretical concepts.

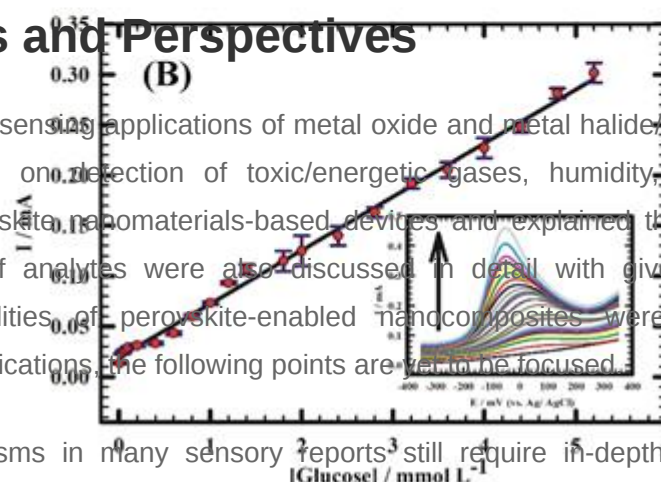


Figure 6. (A) Amperometric response of graphite/Sr₂Pd_{0.7}Au_{0.3}O₃ with successive additions of glucose from 0.2 to 100 μM . Insets (1, 2): calibration curves for glucose for concentrations from 0.4 to 10 μM and from 20 to 100 μM , respectively. We used 0.1 M NaOH and an applied potential of -76 mV. (B) Calibration curve for glucose in diluted urine for concentrations from (10–5.2 μM). Inset: Linear sweep voltammetry (LSVs) of 10 mL of diluted urine at the majority of the device-based sensory investigations are influenced by operational temperature, therefore, considerate optimization is needed to attain responses at room temperature. (reproduced with the permission from reference [121]).

Similar to effective and reproducible and standardize procedure, is required to detect the presence of nitro explosives specifically target (toxic drugs, SnO₂ perovskite oxide (SCO) (Sardar et al., 2014) SnO₂ perovskite oxide was synthesized by the sol-gel method (assisted by citric acid and cetyltrimethylammonium bromide (CTAB) in water) and employed in nitro explosives (TNP, TNT, HMX and RDX) detection in ethyl acetate (EtOAc), tetrahydrofuran (THF) and ethyl methyl ketone (EMK) at room temperature. As displayed in Figure 7, SCO shows better selectivity to nitro explosives than other competitive analytes and solvents. Moreover, it revealed a fast response to all explosives with LODs in the range of 6×10^{-9} to 9.6×10^{-9} M. Regarding the fluorescence-based sensing, this work is an exceptional example, thereby this tactic can be extensively used for other analytes.

- Focus on lead free organometallic halide perovskites is desirable for research in biological imaging studies.
- Research in the development of stable perovskite nanomaterials for environmental and biological assays needs to be intensively stimulated and encouraged.
- Investigations on the incorporation of well-known matrices in perovskite-composite sensors, such as metal organic frameworks, metal nanostructures, hybrid clusters and polymers, need more attention.
- Perovskite nanomaterial-based colorimetric/naked eye analyte determinations require further attention.
- Design and development of low toxic perovskite nanomaterial-based devices/probes towards sensing-drug delivery modules are required to be established in the future.

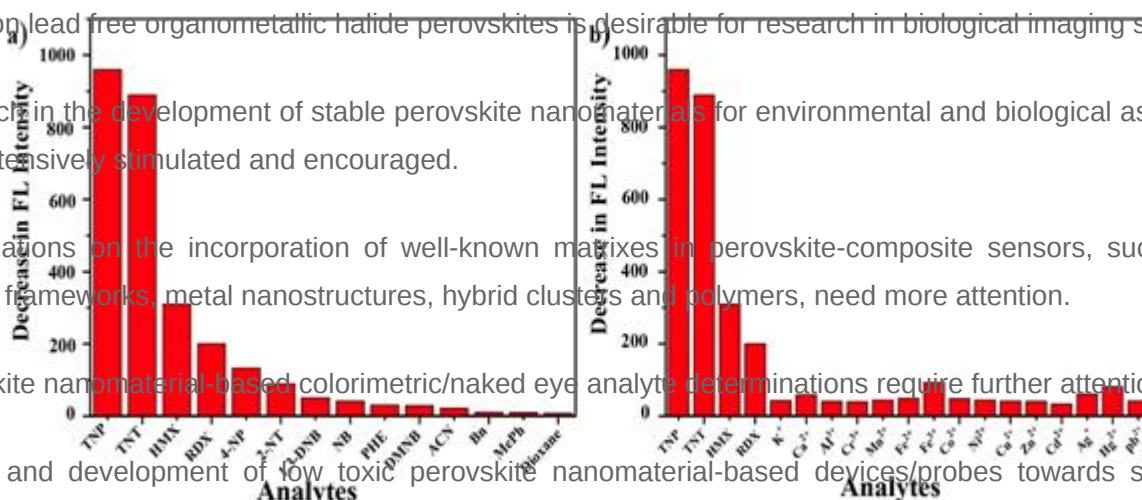


Figure 7. Graphs for selectivity test of SCO in the presence of the (a) nitro compound, aromatic and nonbenzene solvents and (b) various metal ions (the concentration of TNP, TNT, HMX, RDX and the materials used was 30 μ M in SCO (100 ppm), reproduced with the permission from reference [128]).

Next, Hernández Rodríguez et al. described the NMP-doped (1.0% mol) yttrium orthoaluminate nanoperovskites (by the sol-gel method) for temperature sensing in the first and second biological windows (293–611 K), which can be employed in subtissue luminescence imaging [124]. Likewise, nanostructured *p*-type LaCoO₃ was synthesized by solution polymerization tactics and engaged in ultraviolet detection [125]. When exposed to a UV light source, the material showed changes in resistance. These results further confirm the exceptional analytical applications of metal oxide perovskites.

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7. Aamir and coworkers proposed the cesium copper bromide (CsCuBr₃ by wet chemical method) perovskites for aqueous nitro compound detection [128].
8. Aamir and coworkers proposed the cesium copper bromide (CsCuBr₃ by wet chemical method) perovskites for aqueous nitro compound detection [128].
9. Aamir and coworkers proposed the cesium copper bromide (CsCuBr₃ by wet chemical method) perovskites for aqueous nitro compound detection [128].
10. Aamir and coworkers proposed the cesium copper bromide (CsCuBr₃ by wet chemical method) perovskites for aqueous nitro compound detection [128].

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Towards metal ions discrimination, inorganic halide perovskites were effectively employed with certain promising applications. For instance, Sheng et al. established the metal ions sensing ability of cesium lead halide perovskites

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quantum dots via detecting changes in fluorescence [130]. As shown in Figure 8, CsPbBr₃ quantum dots (QDs)

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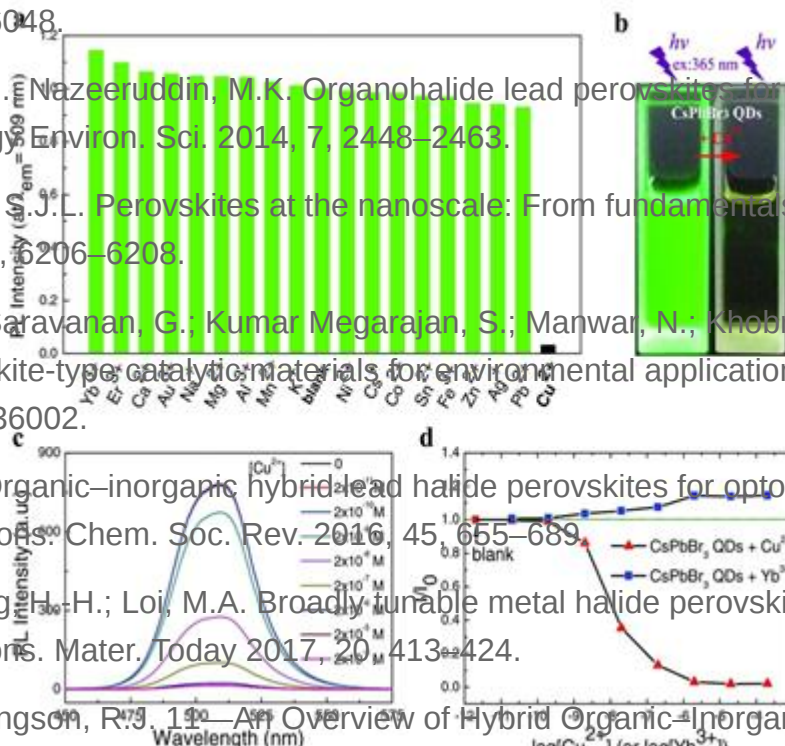
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PL tuned sensing of Cu²⁺ ions was also demonstrated by europium (Eu³⁺)-doped lead free Cs₃Bi₂Br₉ perovskite quantum dots [132]. The Eu³⁺ incorporated Cs₃Bi₂Br₉ QDs with PLQY of 42.4% were prepared from the ligand assisted reprecipitation method and applied in PL based Cu²⁺ sensor. Linear range of Cu²⁺ detection was found to



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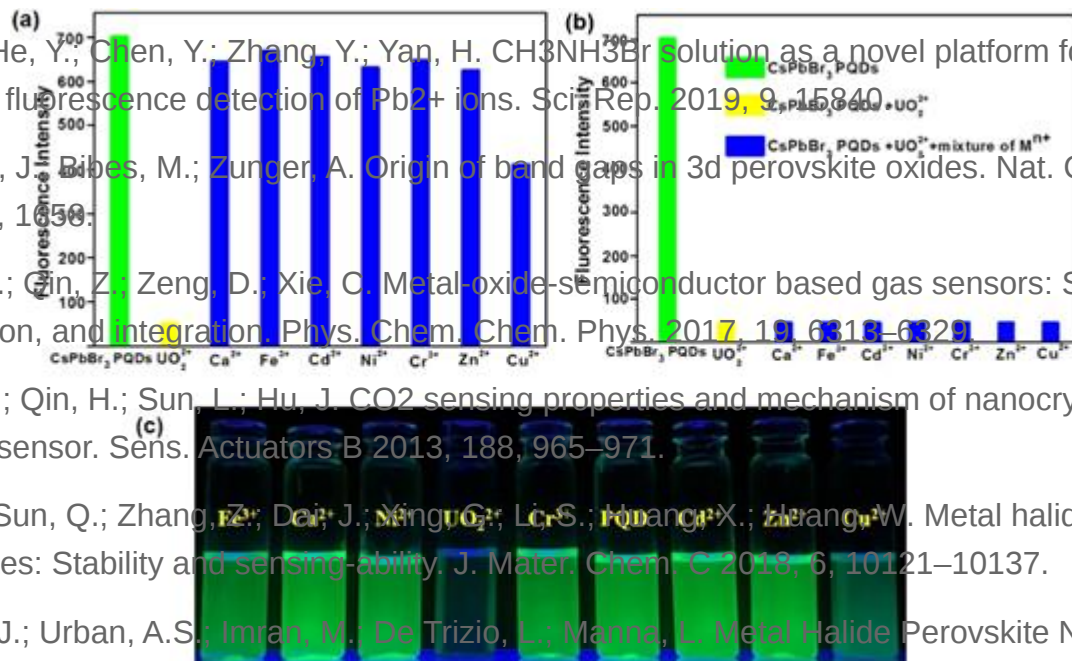
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Curve	Range (ppm)	R ²
1	201.7	0.9999
2	164.9	0.9999
3	154.9	0.9999
4	144.9	0.9999
5	134.9	0.9999
6	124.9	0.9999
7	114.9	0.9999
8	104.9	0.9999
9	94.9	0.9999
10	84.9	0.9999
11	74.9	0.9999
12	64.9	0.9999
13	54.9	0.9999
14	44.9	0.9999
15	34.9	0.9999
16	24.9	0.9999
17	14.9	0.9999
18	4.9	0.9999

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500.0 ppm; 164.0, 148.3, 133.4 (55 ppm); reproduced with the permission from reference (1).

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with position and spin of holes. This can be made clear by substituting the following for (14) in (12):



- Appl. Phys. 2018, 51, 045105. where the conductive holes concentration was enhanced and an increase in the current response was observed. This work has pointed to the exceptional direction in inorganic metal halide perovskite-based sensor research. Next, as illustrated in Figure 12, CsPbBr₃ PODs and CsPbI₃ PODs (prepared by the hot-injection method with PLOY = 52.88% and 46.18%, respectively) display PI quenching to picric acid in solution with estimated LODs of 0.8 nM and 1.9 nM, respectively [143]. In the presence of other competing analytes like 2,4,6-trinitrotoluene (TNT), 2,4-dinitrotoluene (DNT), nitrobenzene (NB), benzoic acid (BA), 1,3-dinitrobenzene (DNB) and benzaldehyde (BD), PODs showed exceptional selectivity to picric acid with inkjet printing applications, thereby they attested as a nice report in the explosive sensor.
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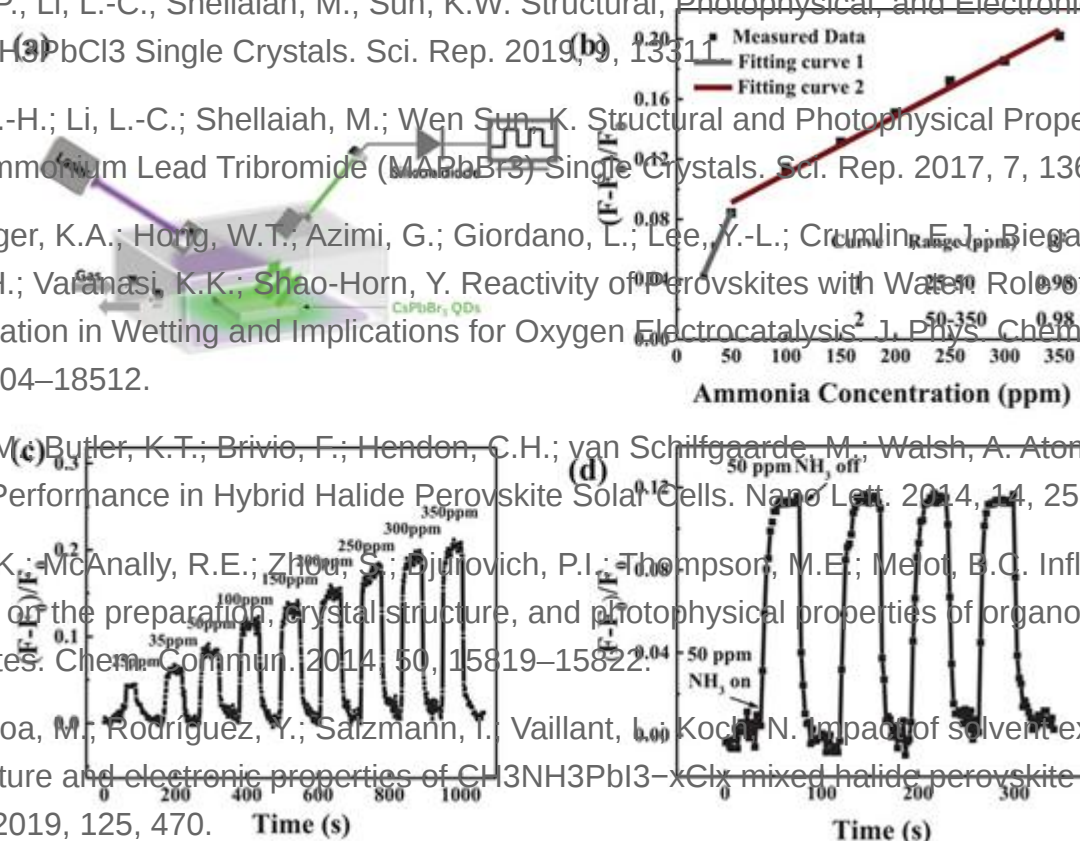
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- Figure 12.** Schematic illustration for the sensitive fluorescence detection of picric acid (PA) based on perovskite quantum dots (reproduced with the permission from reference [143]).
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