

Advancements in ammonia gas detection: a comparative study of sensor technologies

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ABSTRACT

Ammonia gas is a colorless gas that is known for its pungent odor. It is commonly used in various industries, such as agriculture, refrigeration, and chemical manufacturing. This paper provides a comprehensive overview of various technologies employed in ammonia gas sensors. The objective is to compare and identify the optimum method to detect ammonia gas. The review encompasses catalytic gas sensors, metal oxide gas sensors, polymer conductivity gas sensors, optical gas sensors, and indirect gas sensors, detailing their respective operational principles. Additionally, the advantages and disadvantages of each technology for ammonia gas detection are outlined. All these technologies have been used for many applications and some of them have been commercialized. Some sensor characteristics suggestions are also stated in order to develop an improved optical ammonia sensor for industrial applications.

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1. INTRODUCTION

Numerous techniques and technologies are available claiming to possess the capability of ammonia detection. Essentially, ammonia can be effectively detected by the human due to its distinct strong and penetrating odor. The human nose exhibits a high sensitivity to ammonia compared to other odorous gases, even at extremely low concentrations, around 55 parts per million (ppm) [1]. Nevertheless, the human sense of smell falls short in accurately quantifying the quantity of ammonia and detecting the gas at lower concentrations. In various scenarios, it is crucial to determine the ammonia concentration for the sake of health and safety, particularly in the monitoring of ambient air quality [2]. Other industries, such as automotive, chemical, and medical diagnostics, also necessitate the quantification of ammonia for specific purposes outlined by Timmer *et al.* [3]. Consequently, sensors have been developed for the purpose of detecting ammonia gas, enabling the measurement of its concentration, and some of these sensors are capable of detecting remarkably low levels, reaching even parts per billion (ppb) [4]–[6].

There has been extensive research documenting various types of ammonia sensors. This motivates the creation of a paper that consolidates the strengths and weaknesses of current ammonia sensors in one comprehensive review. The paper aims to provide a thorough examination of several ammonia sensors used in general applications. These sensors are classified into five main types, and their respective working

principles, along with their advantages and disadvantages are discussed. They are metal oxide semiconductor sensors, conducting polymer gas sensors, catalytic sensors, indirect gas analyzers and optical gas sensors. Each of these sensor types will be explored in detail to highlight their specific characteristics and applicability.

2. TECHNOLOGY OF AMMONIA GAS SENSOR

Ammonia gas sensor technologies encompass a range of detection methods and offer diverse approaches to accurately detect and quantify ammonia gas concentrations in various applications. Therefore, a review on the different types and technologies of ammonia gas sensors and their working principle will be discussed. The ammonia gas sensor characteristics including their advantages and drawbacks will also be highlighted.

2.1. Metal oxide semiconductor (MOS) sensors

The first category of ammonia gas sensors under discussion employs a metal oxide semiconductor. Typically, these sensors utilize zinc oxide (ZnO) as the metal oxide semiconductor for detecting ammonia [7]–[9]. The high surface-to-volume ratio and distinctive properties of ZnO make it a preferred choice, as these factors significantly impact its gas sensing performance [10]. These gas sensors are compact in size and cost less expensive. This particular gas sensor functions are based on conductance change principle resulting from the chemical adsorption of gas molecules onto the sensing layers. As explained by Rao and Rao [11], the layers of sensing are created by preparing pastes of doped powders of ZnO mixed with distilled water. These pastes are then applied between pre-printed silver electrodes on alumina substrates and air-dried, resulting in the formation of thick-film sensing layers. Rao and Rao [11] also stated that the addition of alumina substrates is intended to evaluate their characteristics as part of the sensing layers.

Four different sensing layers, named ZnO, palladium-zinc oxide (Pd-ZnO), iron-zinc oxide (Fe-ZnO), and ruthenium-zinc oxide (Ru-ZnO), were tested by Rao and Rao [11] to assess their sensitivity. The sensitivity of their sensor was defined by authors as the difference between the voltage readings in the presence of ammonia gas and in the ambient air. The results of this sensitivity evaluation show that the sensitivity increases with higher concentrations of NH₃ for all tested sensing layers. However, the increment is not linear across all the sensing layers examined. The non-linear nature of the graph in these findings can pose challenges for future ammonia quantification processes. Furthermore, there is some doubt regarding the validity of this definition of sensitivity. As per the textbook [12], the sensitivity of a sensor is typically defined as the ratio of the change in output quantity to the corresponding change in the input measurand. To provide an example, let's consider the gas sensor exhibits a correlation where a 100 ppm increase in the input gas concentration results in a corresponding 1 V increase in the output voltage. In this case, the sensitivity of the sensor would be represented as 1/100 V/ppm.

Rao and Rao [11] also conducted an additional trial to showcase the reaction speed of his sensor components when subjected to 30 ppm ammonia in ambient air. The findings indicate that the Pd-ZnO sensor part exhibits a comparatively swift response time of around 4 seconds. Furthermore, the Pd-ZnO sensor element demonstrates the highest sensitivity, as explained by Rao. This superiority can be attributed to the smaller particle size of Pd-ZnO in comparison to the other sensing material.

Extensive research has been conducted on MOS sensors, yielding promising outcomes that highlight their durability and cost-effectiveness [13]–[15]. However, it is important to acknowledge the drawbacks associated with these sensors. One of the primary disadvantages is their lack of selectivity specifically towards ammonia gas [16], [17], especially when operating in the presence of water vapor [18]. For instance, the use of thin tungsten trioxide films to detect ammonia under varying relative humidity levels (2% to 83%) has demonstrated changes in the optical signal [19]. Consequently, addressing this selectivity issue requires the development of a novel approach or method.

Several approaches have been documented to address and mitigate the selectivity problem encountered in MOS sensors. These approaches include the utilization of neural network methods [20], mathematical modelling techniques [21], and the incorporation of specific material or additives that improve the chemisorption of targeted gases [22], [23]. Additional techniques to improve sensor selectivity involve the implementation of masks, filters, or special parameter programming. The programming involves carefully controlling the sensor's temperature to enhance sensitivity towards a specific gas while minimizing interference from other gases [18]. Despite advancements, MOS sensors still face certain challenges. They can be susceptible to electromagnetic interference, and their performance may be influenced by humidity levels. Additionally, MOS sensors often have a limited lifespan due to weaknesses in the filters they employ. Moreover, the complexity involved in fabricating enhanced MOS sensors can result in increased production costs.

2.2. Catalytic sensors for ammonia gas

The catalytic ammonia sensor represents another type of technology for ammonia sensing, which is a variant of the MOS sensor. MOS sensors lack selectivity towards specific gases as elaborated and reported by Aslam *et al.* [18]. To address this limitation, several methods have been utilized to enhance selectivity, including the use of dopants and additives. In the case of catalytic ammonia sensors, catalytic metals are introduced and integrated into the MOS structure, setting them apart from conventional MOS sensors. Material for instance Platinum (Pt), Palladium (Pd), or Iridium (Ir) are employed as gates within the MOS structure to enhance its selectivity abilities [24]. The operation of a catalytic ammonia sensor is based on the behavior of charged particles within the MOS structure. As explained by Winqvist *et al.* [25], when the catalytic metal is exposed to ammonia, its molecules undergo dissociation, and a few of the hydrogen atoms present are captivated at the interface of the oxide material. The presence of absorbed hydrogen atoms results in the formation of dipoles, causing a voltage decrease across the metal oxide interface. As a result, this voltage modification influences the electrical characteristics of the MOS structure or the number of charge carriers along the voltage axis. By employing a field-effect component like a transistor or capacitor, the variation in charge carriers can be detected.

In a research study, the capacitance of the MOS structure was measured by Spetz *et al.* [24] using a Boonton capacitance meter. The meter was operating at 1 MHz and the metal gate used in the experiment consisted of a thick contact dot made of either aluminum or palladium as illustrated in Figure 1. Furthermore, a thin film of platinum was deposited onto the thick contact through the process of evaporation. The result reported that the capacitance is altered when exposed to different amount of NH_3 . This shows that this setup is capable to quantify NH_3 gas. However, this method still encounters some of the drawbacks mentioned earlier, such as interference to other gases. Kim *et al.* [26] has reported that this type of sensor has cross sensitivity with methane gas. The high surface-to-volume ratio and distinctive properties of ZnO make it a preferred choice, as these factors significantly impact its gas sensing performance [10]. These gas sensors are compact in size and cost less expensive.

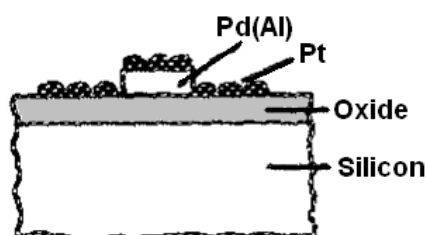


Figure 1. Structure of catalytic metal oxide sensor

In a separate experiment conducted by Spetz *et al.* [24], the changes in voltage that is caused by ammonia was monitored at a constant capacitance. Seven disbursement of ammonia gas with varying concentrations were introduced onto the sensing layer at maintained temperature around $150\text{ }^\circ\text{C}$. The outcome of the experiment shows that the voltage shift becomes higher with respect to higher concentration of ammonia.

One drawback of this sensor system is its susceptibility to electrical interference. The measurement reading is influenced by the presence of charge carriers that are sensitive to electrical fields. Additionally, the sensor described by Aslam *et al.* [18] exhibits a slow response time (more than 10 s). It also shows slow recovery time (more than 900 s). Moreover, the catalytic sensor requires precise temperature control ($300\text{ }^\circ\text{C}$) to ensure selectivity for the desired gas. As a result, this type of sensor may not be suitable for use in many industrial applications that need fast response and recovery time. Furthermore, they are not suitable to be used for gas flows that involve temperature fluctuations, such as in a car exhaust system.

2.3. Conducting polymer gas sensors

Another type of ammonia gas sensor utilizes polyaniline as the conducting polymer [27], [28]. The working principle of this sensor relies on the electrical conductivity of the polymer. The advantages of using polyaniline, including higher sensitivity towards ammonia has been highlighted by Matsuguchi *et al.* [29]. It also has a lower detectable limit (10 ppm), and ability to operate at or near room temperature. Furthermore, polyaniline exhibits high stability against ambient oxidation. It also has reproducibility characteristics when stored in water or air [30]. An improved version of this sensor can achieve even better detection limit, down to one part per million as reported by Chabukswar *et al.* [31].

Kukla *et al.* [28] have reported a conducting polymer ammonia gas sensor. It is based on polyaniline, which is integrated onto a small silicon chip measuring 5 mm by 12 mm. In their study, they conducted measurements of the lateral electrical conductance of polyaniline films with varying thicknesses in an ambient air environment, showing a linear dependence between conductance and film thickness. Additionally, Kukla *et al.* [28] have studied the relation between the resistance and temperature for different concentration of ammonia exposure. It shows that the sensor resistance is temperature-dependent and does not follow a linear trend. Therefore, it needs a signal conditioning circuit to linearize the reading. A structure example of ammonia sensor which has a polyaniline sensing film and signal conditioning circuit is shown in Figure 2.

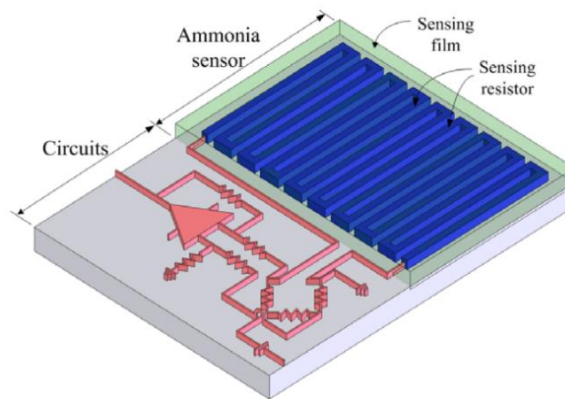


Figure 2. Ammonia sensor structure based on polyaniline

Kukla *et al.* [28] have provided insights into the response time of the polyaniline-based sensor. They noted that the sensor's response is affected by the infiltration of ammonia molecules into the responsive coating, which impacts its performance. Consequently, a thinner polyaniline film was considered preferable. However, it was observed that thinner films had the potential to cause breakage at the metal electrodes. As a result, an optimum layer thickness of 2 μm was utilized. The response and recovery times for various concentrations of ammonia are also displayed in a resistance versus time graph. From the reported graph, it shows a minute of response time and it takes around 2 minutes to recover. It is important to note that using polyaniline as the sensing material has some drawbacks, including a relatively slow response time of approximately 1 minute and its dependence on temperature.

In previous studies [32], [33], polypyrrole served as the polymer film, inducing an alteration in the material's conductivity and presenting itself as a viable option for resistance measurement [34]. As stated by Nylander *et al.* [35], the conductivity of the material reduces when exposed to ammonia. Nevertheless, one disadvantage is that it experiences a partially irreversible reaction when exposed to relatively low levels of ammonia which is less than 108 ppm [36]. The chemical reaction involving polypyrrole and ammonia can only be partially reversed through purging with nitrogen or air. Consequently, this leads to an accumulation of mass in the polymer film, reducing the sensor's sensitivity over time [33]. Moreover, the extended exposure of polypyrrole film to moist ammonia over a prolonged duration (7 days) results in a fully irreversible reaction [36]. In conclusion, these two materials (polyaniline and polypyrrole), when employed as conducting polymers for ammonia sensing, possess their own individual limitations.

2.4. Indirect gas analyzer

According to the findings in these two journal reports [18], [37], a majority of the existing ammonia sensors exhibit limited selectivity towards ammonia when compared to other gases. Therefore, a solution to cater this issue was initiated. A new setup for gas sensor system was introduced where it integrates a filtration component that enables only the desired gas to be introduced to the system's detector. A method that involves gas diffusion separation using special membranes that can permeate the gas was introduced by Timmer *et al.* [38]. They conducted a test using polytetrafluoroethylene (PTFE) and hydrophobic polypropylene (PP) membranes to detect ammonia. The sensor system consists of three main components as shown in Figure 3.

The first part is the gas sampler, where the air and ammonia were introduced. The second part is called the selector where only ammonia is allowed to pass through, and the last part is the detector. The gas sampler and the selector components comprise two parallel channels that are separated by a membrane that

can repel water. Timmer *et al.* [38] conducted two tests to evaluate the performance of the sensor setup. In the first experiment, the selectivity of the system was examined. Observations revealed that the system exhibited no response to typical ambient CO₂ concentration levels, indicating that the gas-permeable membrane effectively blocked unwanted gases. The second experiment focused on the sensitivity of the sensor system. Different concentrations of ammonia that was diluted using nitrogen gas were used to test the sensitivity. Initially, a higher ammonia concentration of 9.8 ppm was employed. Then, the ammonia level was gradually diluted and conductivity readings were recorded. The result is reported as the conductivity is proportionally increased with ammonia concentration. However, the result shows a minus point where the increment is not linear.

Timmer *et al.* [38] present an ammonia sensor capable of detecting low ammonia concentrations as low as 0.3 ppm. However, the study also highlights a few drawbacks of the system. One disadvantage is the potential of gas bubbles become trapped inside the electrochemical (EC) detector, which can lead to a conductivity offset and reduce the system's reproducibility. Additionally, the system exhibits a slow response time, with the testing gas taking approximately 10 minutes to reach the detector. The use of membranes for gas separation makes the sensor less robust as compared to other ammonia sensor systems, as clogging issues tend to arise over time. This aspect is the main concern for those who plan to measure ammonia in hazy environment, where dust and heavy particles may be exist.

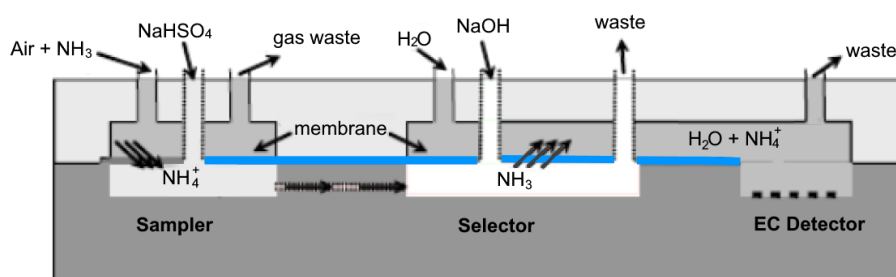


Figure 3. Ammonia sensor using membrane as selective mechanism

2.5. Optical gas sensors

An optical gas sensor is a device designed to detect and measure the concentration of gas using optical properties such as using refractive index changes [39]. For ammonia gas sensing, it operates based on the interaction between ammonia molecules and specific sensing materials or indicators [40], [41], which produce a measurable optical response. Typically, the sensor utilizes a light source, such as a laser or LED, to emit light that interacts with the sensing material. The presence of ammonia alters the optical properties of the sensing material, resulting in changes in light absorption, reflection, or emission [42]. These variations are then detected and converted into an electrical signal, providing quantitative or qualitative information about the ammonia concentration. Mostly, optical sensors employ optical fibers cable to transmit or received signals [43], [44]. Optical ammonia sensors offer advantages such as high sensitivity, real-time monitoring capabilities, and the potential for remote or distributed sensing applications [45].

A common example of the ammonia optical sensor is the research that was conducted by Kalvoda *et al.* [46]. The optical sensor system typically comprises a light source and a detector such as a spectrometer as depicted in Figure 4. The testing gas chamber facilitates the interaction between the measured substance and the light. Kalvoda *et al.* [46] introduced a plasmonic nanostructured array designed specifically for ammonia gas sensing. It utilizes a combination of fiber optics, the extraordinary optical transmission (EOT) effect, and a chemo-optical transducer with selective sensitivity to ammonia gas. The research involved analyzing the spectral transmission of the resulting structure and its alterations when exposed to ammonia gas at various concentrations. Their findings revealed distinct changes in the ammonia spectrum within the Vis-NIR wavelength range, with the most pronounced transmittance changes occurring at approximately 350 nm and 480 nm.

In the laboratory, a series of experiments were conducted to characterize the sensor system. The dilution of the ammonia gas was achieved through the utilization of mass flow controllers (MFC), which provide an absolute accuracy of around one percent. The measurements obtained from the sensor setup exhibited a favorable alignment with the known concentrations of ammonia, demonstrating the system's reliability and accuracy. Kalvoda *et al.* [46] reported a high accuracy ammonia sensing system except for measurements at the very low concentration. The authors deemed this analytic sensitivity or detection limit to

be adequate for ammonia detection in open environments. However, it is worth noting that the experimental setup was placed in laboratory environment where interferences with other gases may not occur. The potential cross-sensitivity with water moisture and CO₂ for open air ammonia measurement is a big concern.

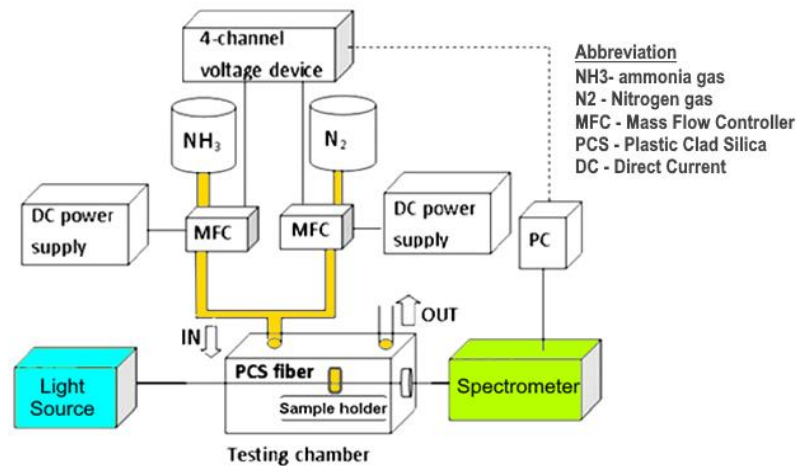


Figure 4. Optical sensor block diagram

There is also another example of optical ammonia sensor that operates by utilizing the photoacoustic effect. In this process, a specific wavelength of light, often emitted by a laser, interacts with the targeted gas molecules, causing them to absorb the light energy and release heat. This leads to the generation of sound waves, which are then detected by a microphone [47], diode laser [48], interferometer [49] or other acoustic spectroscopy sensors [50], [51]. By analyzing these sound waves, the concentration of ammonia gas in the environment can be determined. One advantage of photoacoustic technology is its remarkable detection range, which can extend to as low as 0.1 ppb [51]. However, the photoacoustic sensor also has their own disadvantage. Its complexity and the requirement for sophisticated instrumentation, which may make it relatively costly compared to some other sensing techniques. Additionally, the setup and calibration of photoacoustic systems can be intricate, requiring expertise in optics and acoustics. The catalytic ammonia sensor represents another type of technology for ammonia sensing, which is a variant of the MOS sensor. MOS sensors lack selectivity towards specific gases as elaborated and reported by Aslam *et al.* [18]. To address this limitation, several methods have been utilized to enhance selectivity, including the use of dopants and additives.

2.6. Other types of ammonia gas sensor

Another example of ammonia gas sensing approach involves the use of a chemical reagent that undergoes a color change in response to ammonia. pH paper is a good example of this, where the coloration reaction is subject to the amount of ammonia. The range of the measured gas can be altered based on the coloration reaction. The Nessler reaction [52] is a commonly used coloration reaction for dissolved ammonia, and it has been commercialized. It is used to determine ammonia concentration in solution. However, it should be noted that this reagent is poisonous and it is not appropriate to be used frequently. Additionally, it can be influenced by other gases such as SO₂, which is acidic and can potentially impact the color change of the pH paper. While the sensor system exhibits advanced features, its intricate design contributes to significant manufacturing expenses, thereby reducing its feasibility for widespread practical applications.

There is also other technique known as calorimetry method. It can be used to measure ammonia concentration indirectly by determining the heat released or absorbed during reactions involving ammonia. One common method is the use of an ammonia-selective ion electrode combined with a flow calorimeter or isothermal calorimeter. In this method, a sample of air containing ammonia gas is mixed with a known amount of an absorbent solution, such as sulfuric acid or hydrochloric acid, in a calorimeter. The absorption of ammonia gas by the solution is an exothermic reaction, resulting in a change in temperature that is proportional to the amount of ammonia gas present [53].

The main advantage of using calorimetry method is that it is highly sensitive to heat and can detect low concentrations of ammonia in real time. However, it has very limited range as compared to other analytical techniques, particularly for extremely low or high concentrations of ammonia. Even though

calorimetry offers advantages such as high sensitivity and real-time monitoring, it also has limitations related to calibration requirements, complexity, and range of measurement. The suitability of calorimetry for measuring ammonia concentration depends on the specific application requirements and the available resources.

3. SUMMARY

There are many types of ammonia sensors and their technologies has been discussed in this paper. In order to compare and see the differences clearly, it is summarized and placed in Table 1. This table provides a comprehensive overview of the various ammonia gas sensor including their working principles, advantages, and disadvantages.

Table 1. Ammonia sensors technologies

Sensor type	Working principle	Advantages	Disadvantages
Metal Oxide semiconductor sensors	Utilize metal oxide semiconductor (e.g., ZnO) for gas detection based on conductance change from gas adsorption	Compact, cost-effective, high sensitivity	Lack of selectivity, susceptibility to interference, limited lifespan
Catalytic sensors	Incorporate catalytic metals (e.g., Pt, Pd) into MOS structure for enhanced selectivity	Improved selectivity, enhanced sensitivity	Susceptibility to electrical interference, slow response and recovery time
Conducting polymer gas sensors	Use conducting polymers (e.g., polyaniline, polypyrrole) based on electrical conductivity for gas detection	Higher sensitivity, lower detectable limit, stability	Relatively slow response time, temperature dependence, partially irreversible reactions
Indirect gas sensors	Measure ammonia indirectly through gas diffusion separation using membranes and color change reactions	Simple, cost-effective, indirect measurement	Limited range, potential interference from other gases
Optical sensors	Operate based on light absorption, reflection, or emission principles to detect and measure ammonia gas concentrations	High sensitivity, real-time monitoring, remote sensing capabilities	Complexity in calibration and setup, costliness
Others	Utilize color change reactions (e.g., Nessler reaction) to detect ammonia	Simple, cost-effective	Potential interference from other gases, toxicity of reagents

4. CONCLUSION

In this paper, various types of ammonia gas sensors were evaluated for their suitability to be used in real environments. While some of the sensors showed potential, none of them fully addressed all the key characteristics required for an ideal ammonia sensor, such as selectivity, sensitivity, and fast response time. Based on the review, it can be concluded that optical gas sensors, which utilize light absorption technique has the potential to meet the requirements of a good ammonia sensor. Therefore, it is strongly recommended that any new development in the field of ammonia sensors should focus on the optical principles if budget is not a constraint. However, it is important to address the limitations of existing optical ammonia sensors, such as their cross sensitivity to water vapor, oxygen and carbon dioxide gas, as well as their slow response time. These issues need to be addressed in order to develop an improved optical ammonia sensor for industrial applications.

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