Volatile Organic Compound Sensing Array and Optoelectronic Filter System using Ion-Pairing Dyes with a Wide Visible Spectrum

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An ideal dye-based sensing array has essential design requirements, including facile preparation methodology, tolerance to water vapor, a broad range of color-responsive changes, and a simple readout system. Here, a brief synthetic route is developed for ion-pairing dyes exhibiting unusual chromatic changes across the entire visible spectrum. It requires only mixing and precipitation under mild conditions. The dyes are applied to a sensing array containing 12 sensing elements with different initial states. Owing to the numerous color variations of the dyes, the color map generated by the array is highly simple yet sufficiently accurate to distinguish among the different functional groups (such as amines, aldehydes, and carboxylic acids) as well as carbon chain lengths. Principle component analysis (PCA) demonstrates that volatile organic compounds (VOCs) can be well classified according to the color changes of the sensing array. The ion-pairing dyes are embedded into 3D stacked nanofibers via electrospinning, and function as effective harmful-gas (e.g., formaldehyde) sensors with sub-ppm theoretical detection limits (0.15 ppm). Finally, the 3D stacked nanofibers can be employed in an optoelectronic filter system that automatically checks for formaldehyde in the surroundings and also confirms the effective removal of the detected formaldehyde by the gas filter cartridge.

1. Introduction

Air pollution is one of the five leading contributors to global health risks, the others being diabetes mellitus, high blood pressure, obesity, and tobacco smoking.[1,2] Globally, nearly 80% of people breathe air containing hazardous pollutants whose concentrations exceed the recommended standards stipulated by the World Health Organization.[3] Air pollution is caused by the presence of several toxic materials, among which VOCs are an important group of industrial pollutants.[4] However, it is difficult for human sensory organs to detect VOCs, which is highly concerning as VOCs have a significant negative impact on human health.[5,6] Therefore, with progress in research on monitoring VOCs in the surroundings, there is a demand for facile, sensitive, convenient, and cost-effective analytical technologies. In this regard, colorimetric sensors based on the optical properties of colorants have been investigated as effective tools for the visual detection and discrimination of diverse vapors.[7–10] This research topic can be primarily classified into “sensing materials” and “detection methodologies.”

The intensive development of “sensing materials” has yielded many useful sensors based on various intermolecular interactions such as coordination, charge transfer, acid–base interaction, and π–π molecular complexation.[11–15] Numerous sensors based on these mechanisms have been successful in detecting and distinguishing VOCs. In most of these sensors, chemoresponsive dyes have been used as the sensing material because various sensing systems with dyes have shown chromatic responses to vapors in the solid-state.[16–18] Because most dyes are sensitive to large environmental changes in humidity, minimizing the water vapor interference is considered an important challenge.[19,20] Any significant response to ambient humidity is especially detrimental to the sensor performance, leading to the poor detection of target analytes at low concentrations. To minimize dye–humidity interactions, hydrophobic materials must be used as either matrices or substrates, which has significantly restricted the scope of available dyes.[21] Controlling the color changes is another major consideration while designing chemoresponsive dyes as sensing materials. Previous routes to synthesize dyes with controlled color changes have employed diverse strategies for structural change by considering the functional groups, carbon-chain lengths, and metal-ligating sites.[22–24] These have facilitated an alternate means to regulate the chemical sensitivity and selectivity of the dyes to specific...
targets; however, their synthesis procedures are sophisticated, time-consuming, and require trained professionals. Further, the dynamic color range generally comprises only two different colors and still falls within the narrow visible spectrum, that is, the range of color change in typical dyes is narrow.

In the field of “detection methodologies,” array sensing platforms based on the optical properties of sensing materials are considered a powerful tool because of their intelligence in visually identifying and distinguishing various VOCs. Based on cross-reactive sensory elements, these systems chiefly mimic the human olfactory system by generating composite responses as a unique fingerprint for each VOC. These platforms typically consist of many sensing materials as cross-reactive elements because they require various chemical interactions, such as those related to redox reactions, local polarities, Lewis acids/bases, and Brønsted–Lowry acids/bases. However, the presence of numerous components to produce unique fingerprints further complicates and impedes the interpretation of array responses. These complex readout systems require additional devices to generate identifiable signals from the composite responses, which otherwise cannot be perceived visually. Alternatively, using target-specific responsive chemicals enhances the simplicity and sensitivity of the detection of certain vapors, similar to that in an enzyme-substrate reaction. However, the data obtained by the use of such chemicals have shown that the developed array only discriminates structurally similar targets with identical functional groups, necessitating the design of a simple but widely applicable detection methodology.

A common theme in the research on colorimetric sensors is the concurrent development of chemoresponsive dyes (sensing materials) and array sensing platforms (detection methodologies). Therefore, an ideal sensing platform with chemoresponsive dyes to visualize VOCs should possess the following features. First, the dyes should be tolerant to water vapor (i.e., be hydrophobic), and their dynamic color range should belong to a wide visible spectrum. Second, the sensing array should generate a simple, sensitive, and unambiguous composite response without requiring numerous dyes. Third, the sensors should be able to identify various VOCs and provide useful and visually perceptible information.

Considering all the aforementioned limitations, herein, we report a brief, two-step synthetic procedure for ion-pairing hydrophobic dyes that exhibit color changes over a broad visible spectrum. The ion-pairing dyes exhibit significantly more color variations than others and facilitate color tunability through ion-pair-composition variations without the need for complex processes. We fabricated an array sensing platform composed of only three ion-pairing dyes with different initial states. Compared to previous studies on vapor detection, our sensing array produced a simple color pattern enabling qualitative discrimination among various VOCs with different alkyl chain lengths, thus eliminating the requirement of additional devices, such as image processors, potentiometers, resistance meters, and interdigitated transducers. Furthermore, using electrospinning, we fabricated colorimetric 3D stacked nanofibers embedded with the ion-pairing dyes as an effective formaldehyde gas sensor. Owing to their desired characteristics such as flexibility, breathability, and gas diffusivity, it is possible to develop optoelectronic filter systems with 3D stacked nanofibers to generate signals containing information of practical significance to workers, such as the filter performance level, optimal replacement time, and environmental information. Therefore, the appropriate course of action under a given circumstance can be easily established according to the electronically triggered color signatures of light-emitting diodes (LEDs).

2. Results and Discussion

Our design strategy involves the formation of an ion pair consisting of charged hydrophilic dyes to widen the color-change range and alter the solubility properties without any chemical modification. A typical procedure for the synthesis of ion-pairing dyes is presented schematically in Figure 1a. After screening the potential dye candidates, we employed five water-soluble charged dyes, viz., crystal violet (CV), brilliant green (BG), ethyl orange (EO), methyl red (MR), and α-naphthol orange (NO), as shown in Figure 1b–d. The dyes were selected on the basis of their coloration, pH range, and ion-pairing interactions. As shown in Figure 1a, heterogeneous mixtures are obtained immediately owing to the formation of water-insoluble complexes. The resultant decrease in the water solubility is attributed to the masking of the originally hydrophilic surface area by hydrophobic domains, such as aliphatic or aromatic groups. Therefore, the ion-pairing dyes aggregate and precipitate in the aqueous media.

Figure S2, Supporting Information, shows the results of the Fourier transform infrared (FTIR) spectroscopy analysis of the ion-pairing dyes. As hydrogen sulfate (HSO$_4^-$) is ionically bonded with BG, the absorption bands of S=O and S−O are observed at 1184 and 999 cm$^{-1}$, respectively (Figure S2b, Supporting Information). However, these bands are not observed in the IR spectrum of BG–MR, indicating that hydrogen sulfate is replaced by another anionic ion, such as that from an anionic dye. Therefore, the FTIR analysis suggests that the oppositely charged dyes are attracted to each other by intermolecular coulomb force, which hold them together as a single molecule. These ion-pairing processes involve stoichiometric association between the cationic and anionic dyes, as confirmed by proton nuclear magnetic resonance (1H NMR) spectroscopy, especially CV–EO = 1:1, BG–MR = 1:2, and BG–NO = 1:1 (Figure S3, Supporting Information). To demonstrate the color changes with changes in pH, solutions containing the ion-pairing dyes at diverse pH levels (ranging from pH 1 to 13) were prepared and photographed; the photographs are shown in Figure 1e–g. The color change in the dye solutions is visually observable throughout the pH range from acid to alkali. For instance, the CV–EO solutions change from colorless to violet in the pH range of 1–5 and to intense yellow at pH > 12. Further, the absorbance spectra of these solutions over the same pH range were recorded and converted into the x and y values of the International Commission of Illumination (CIE) color space to qualitatively analyze the pH response (Figure 1h–j, Table S1, Supporting Information). The color changes correspond to changes in the x and y values. The large changes in x and y yield a C-shaped curve that spans a wide color space depending on the pH levels. For CV–EO, the lowest x and y were observed
at pH 2 (0.233 and 0.222, respectively). There was a gradual increase in $y$ with increasing pH levels. On the other hand, $x$ decreased until pH 6, and then increased with increasing pH levels. As the pH increased and reached 13, $x$ and $y$ demonstrated the maximum values of 0.400 and 0.391, respectively. Similarly, BG–MR and BG–NO also showed different $x$ and $y$ values with changes in pH levels. Thus, these quantitative analyses made it possible to distinguish pH levels that are otherwise difficult to identify visually. The bathochromic and hypsochromic shifts were established by comparing the molecular-scale structures of the ion-pairing dyes, and the results indicated that the cationic and anionic dyes do not lose their optical properties when forming ion-pairing dyes, thereby leading to a wide pH response range (Figures S4–S6, Supporting Information).

When compared to previous studies, our suggested procedure for synthesizing ion-pairing dyes under various pH conditions and developed a sensing array consisting of three dyes with different initial states. Our sensing array was highly simple but could visually discriminate chemically diverse VOCs (Figure 2a). The Euclidean distances (EDs, defined as the square root of the sum of different RGB values) and Hue values (representing a dimension of coloration) of the sensing array were calculated and are presented in Figure 2b–d and Table S3, Supporting Information, respectively.[47,48] To distinguish aldehydes from carboxylic acids, we incorporated a tertiary amine (3° amine), 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU), into the sensing array, thereby affording different color response profiles upon exposure to different carbonyl groups (Figure 2a,c,d). Because 3° amines have no hydrogen bonded to the nitrogen, they do not form stable derivatives with aldehydes.[49] Therefore, there are no color changes for the 3° amine upon exposure to aldehydes. However, since carboxylic acids have no selectivity to amines, large color changes are observed for both alkaline
states. For carboxylic acid groups, the EDs generally decreased with an increase in the carbon chain length (Figure 2d). We deduced that this tendency originates from the difference in volatility and penetration into the polymeric matrix depending on the molecular weight of vapors, and thus can be employed to distinguish vapors with different carbon numbers in the same functional groups. The color changes in the array were equal for identical dyes after ammonia exposure, irrespective
of whether the original state of the dyes was acidic or alkaline (Figure 2a). However, most ion-pairing dyes displayed different coloration to each VOC when their initial states differed. Therefore, based on the EDs, we could obtain contour plots of the sensing array for representing unique patterns of different VOCs in a 2D format (Figure S7, Supporting Information). This allowed us to qualitatively visualize the color changes of the array and identify VOCs through colorimetric patterns as a fingerprint. These resultant color changes demonstrated that ion-pairing dyes interact uniquely with each VOC, resulting in discriminable responses upon vapor exposure. These variations in the colorimetric response are evidence that the initial state of ion-pairing dyes is the major factor affecting the degree of color change, which further suggests that the dyes exhibit another chromatic behavior as well as halochromism (for details, see Figure 3). Owing to these multiple optical properties, despite

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**Figure 3.** Chromatic dynamics of the colorimetric sensing array for different carboxylic acids. a) Time-lapse photographs of all 12 sensing elements upon exposure to acetic acid, butyric acid, and valeric acid. b–d) Corresponding ED curves calculated from the extracted RGB values using their time-lapse photographs.
the fact that the sensing array in this study is composed of only three ion-pairing dyes with different initial states, a simple and unique differential color map can be produced, analogous to human fingerprinting, without requiring numerous colorimetric dyes and additional devices (as listed in Table S4, Supporting Information). To explore the differences between the functional groups of VOCs, we performed a PCA, which generates a linear combination of the EDs dataset collected from five independent experiments (Figure 2e–g). The PCA results generally require three or four dimensions to capture >95% of the total variance for a precise classification (Figure 2e). To simplify the plots and aid in visualization, we only employed the first two principal components that captured >80% of the total variance for the total classification. As shown in Figure 2f, five repetitions of individual datasets show good clustering and resolution without any ambiguity. They are also classified into distinct clusters without any overlap, even for similar chemical structures with a difference of one or two carbon numbers (Figure 2g), that is, carboxylic acids with different numbers of carbons can be differentiated.

The temporal response of the sensing array to VOCs was also investigated to determine the chromatic behavior of the ion-pairing dyes (Figures S8–S11, Supporting Information). Figure 3 shows the variations in color changes and EDs for diverse carboxylic acids; the time-lapse photographs were captured at intervals of 0.5, 5, and 10 min up to 6, 60, and 120 min, in that order, for acetic acid, butyric acid, and valeric acid, respectively. As shown in Figure 3a, the color-change rate differs significantly depending on the length of the carbon chain. Therefore, the carboxylic acids are accurately distinguishable by monitoring the color-change trends, without any qualitative and statistical analysis such as those involving the ED and PCA. For the default state, BG–MR exposed to acetic acid produces a green color within 6 min, whereas that exposed to butyric acid exhibits a green color within 35 min. Since carboxylic acids with short carbon chains have a high volatility and low acid dissociation constant (pK_a), the vapor causes large changes in the pH and easily penetrates the polymeric matrices of the sensing array. Therefore, both, the rate and degree of color changes (i.e., the ED) increase with decreasing carbon number of the carboxylic acid. The overall color changes are faster and larger for the primary amine (1° amine), 2,2′-(ethylenedioxy) bis(ethylenimine) (EDEA), than for the 3° amine, implying the higher reactivity of the 1° amine than the 3° amine toward a carboxylic acid due to steric and electronic effects (Figure 3b–d).

In contrast, the use of valeric acid results in more significant deep-green transformation in acidic state BG–NO than other carboxylic acids with a lower pK_a, as shown in Figure 3a. In addition, the coloration of BG–NO (for the 3° amine) changes from pink to yellow to green, and not from pink to blue to green, with decreasing pH levels (Figures 1g and 3a); this suggests that the color changes of BG–NO (for the 3° amine) upon exposure to carboxylic acids are different from those with changes in pH levels. Therefore, we can deduce that the color changes originate not only from variations in the acidity/alkalinity of VOCs but also from an intrinsic solvatochromic effect of the ion-pairing dyes. As shown in Figures S8–S11, Supporting Information, not all dyes exhibit coloration corresponding to their acidities owing to the dominance of solvatochromism to halochromism in such dyes. Thus, the ion-pairing dyes have two contrasting chromatic behaviors, allowing them to exhibit a wide variety of colorimetric responses to chemically different VOCs.

To design an effective harmful-gas (e.g., formaldehyde) sensor, we prepared 3D stacked nanofibers by electrospinning to obtain a high surface area, porosity, and large space between the fibers (Table S5 and Figure S12, Supporting Information). Thus, we could impart the desired characteristics such as flexibility, breathability, and gas diffusivity into the nanofibers. Figure 4a presents a schematic of our preparation process; we chose CV–EO with the 1° amine as the sensing material for formaldehyde detection owing to its large ED and fast response to aldehyde groups (Figure 2c and Figure S10, Supporting Information). The photograph of the 3D stacked nanofibers in Figure 4b shows a uniform light-yellow coloration of the nanofibers without the agglomeration of dyes, indicating that CV–EO is uniformly incorporated within the prepared architectures. Figure 4c shows the morphology of the nanofibers with an average diameter of approximately 131 nm, which facilitates the diffusion of gas molecules into the porous matrices, enabling a high sensitivity and fast response to analytes. On coming in contact with formaldehyde of various concentrations (1–20 ppm), the nanofibers exhibit distinct concentration-dependent color changes from light-yellow to blue-gray. Differential color maps were generated by subtracting their RGB values to better visualize the chromatic response (Figure 4d). These results show that the colorimetric signals change with the formaldehyde concentrations, and that the nanofibers can detect formaldehyde up to a concentration of ∼1 ppm. Quantitative analysis was also performed by monitoring the temporal behavior of the ED. As shown in Figure 4e, the ED reaches different equilibrium points at different formaldehyde levels, enabling the quantitative detection of formaldehyde gas even at low-ppm concentrations. Further, the limit of detection (LOD) of the nanofibers, as defined by the minimum concentration corresponding to the ED, is three times the standard deviation (σ) from the mean of the blank control (Figure 4f). The nanofibers have an LOD of ∼0.15 ppm, which is five orders of magnitude lower than the permissible exposure limit (0.75 ppm).

On the basis of the quantified color changes, the nanofibers can be employed as a colorimetric membrane to develop an optoelectronic filter system that alerts users during hazardous situations (Figure 5a,b). Figure 5a shows a schematic of our developed system, which comprises several parts: RGB sensors, cylindrical filters, colorimetric membranes, and a gas mask with LEDs. The RGB sensors were placed on both sides of the cylindrical filter to monitor the RGB values from the colorimetric membranes of the triplet filter layers. When the membrane color changed upon exposure to formaldehyde gas level that is immediately dangerous to life or health (20 ppm), the LED on the gas mask changed from green to red. Because the two LEDs individually denote the inlet and outlet gas conditions, three different states can be represented as shown in Figure 5c (for details, see Table S6, Supporting Information). Therefore, their color signature indicates the necessary course of action in a given environment (Table 1). For example, in the conditional safe state, the formaldehyde passes through the
inlet membrane and is absorbed into the activated charcoal powder. Thus, the right LED changes to red, while the left LED maintains the initial green color, suggesting that although formaldehyde is present in the surrounding environment, the safety of the user can be ensured by wearing a mask with a suitable filter. Hence, the green–red signal of the LEDs is a recommendation to continue wearing the mask. Similarly, the green–green signal represents a completely safe state with no requirement to wear a mask, while the red–red signal represents a dangerous state in which the user is not protected from the formaldehyde even with the mask on. Figure 5d–f shows the formaldehyde-induced chromatic response of the membrane measured by RGB sensors for the previously described three states. As shown in Figure 5d, the 30 experimental trials are arbitrarily classified into three states, and all of them are correctly diagnosed by this system. In addition, complete grouping is seen for the repetitions of each trial, and sufficient separability exists between the presence and absence of formaldehyde gas (Figure 5e). The kinetics of the chromatic response was also monitored by recording the ED every minute (for the dangerous state, Figure 5f, see Supporting videos). The formaldehyde gas flow continued for 10 min, after which the ED was obtained continuously for another 10 min. The response curves of the inlet and outlet exhibited a stable baseline at first followed by immediate color changes (in only a few minutes), reaching maximum levels after exposure to formaldehyde gas.

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3. Conclusion

We have demonstrated that ion-pair formation is a simple and effective method for the synthesis of sensing materials (i.e., ion-pairing dyes) with desirable characteristics that allow multiple coloration and extension of the dynamic color range. Furthermore, their water solubility decreases by the masking of the originally hydrophilic surface area by hydrophobic
The complexes were purified by centrifugation (5000 revolutions per minute (rpm), for 5 min) to remove the water-soluble moieties (i.e., unpaired dyes). Finally, the precipitate was dried under vacuum at room temperature for 24 h (90%, 93%, and 86% yields for CV–EO, BG–MR, and BG–NO, respectively).

**Fabrication of the 12-Element Sensing Array:** The sensing array was fabricated according to the following protocol. First, a polymer/dye solution was prepared by dissolving 0.05 g of the ion-pairing dye in 10 mL of a 13 wt% PVP solution (a 5:5 v/v mixture of DMF and EtOH). Second, to create different initial states, the prepared solution (10 mL) was mixed with 0.15 mL of either acidic or alkaline chemicals (sulfuric acid for the acidic state, EDEA for the 1° alkaline state, and DBU for the 3° alkaline state); the resultant solution was stirred until uniform coloration (inferred visually). Last, 15 µL of the polymer/dye solution was drop-cast onto a microscope glass slide, and the solvent was allowed to evaporate using a heat gun (HL 1610S, Steinel, Einsiedeln, Switzerland).

**Chromatic Analysis of Vapor-Series Exposure:** The 12-element sensing array was used to perform vapor-sensing measurements using an experimental setup (schematically illustrated in Figure S14, Supporting Information). For the optical test, 5 mL of the VOCs was dropped in the crystallizing dish, and the dish was sealed by covering it with an optical glass plate. Because excess VOCs were added to the sealed dish, the dish was saturated with their vapor pressure at 25 °C upon reaching equilibrium. Subsequently, the sensing array was placed in the dish in which the saturated vapor could be maintained. All eight VOCs were exposed to the sensing array, and quintuplicate arrays were used for individual VOCs to demonstrate the reproducibility of the sensor response. To ensure uniform brightness, the array was illuminated using a handheld LED lamp (18 V XR Handheld, DEWALT, MD, USA). The images were collected and analyzed using a digital camera (IXUS 275 HS, Canon, Tokyo, Japan) and photoediting software (Photoshop CS6, Adobe, CA, USA). The software averaged the RGB values of each center of the element spots (11 by 11 pixels), thereby avoiding spot nonuniformity, especially at their boundaries. Differential color maps were obtained by subtracting the RGB values of the images before and after vapor exposure (RGB values of 3–10 expanded to 0–255), thus enabling visualization of the absolute values of the color changes in response to a series of vapors.

**3D Stacked Nanofibers Fabrication by Electrospinning:** CV–EO (0.015 g) was dissolved in 3 mL of a 13 wt% PVP solution in DMF. The PVP/CV–EO solution was then mixed with 50 µL EDEA and vigorously shaken for 5 min, yielding a clear yellow solution. This precursor solution was electrospun at a high voltage (18.8 kV), and the distance between the tip (24 G, 0.3 mm inner diameter) and the drum collector (50 rpm) was 15 cm. A feed rate of 0.8 mL h⁻¹ was used to produce the 3D stacked nanofibers. During electrospinning, the humidity and temperature were maintained at approximately 35% relative humidity (RH) and 25 °C, respectively. This electrospun nanofibers were then dried under vacuum to remove any residual solvent before use.

**Acquisition of Color-Change Profile for Caeous Formaldehyde:** The prepared nanofibers were used to measure formaldehyde gas-sensing responses. The sensing experiments were conducted using a customized gas instrument (Figure S15, Supporting Information). Mass flow controllers (D-6361-DR for nitrogen and F-201CB for
formaldehyde, Bronkhorst, Gelderland, Netherlands) were briefly used to generate gas streams containing the desired levels of RH and formaldehyde. The gas concentration was verified using a portable air quality monitor (Series 200, Aerogal, Auckland, New Zealand). The RH was maintained at 50% by mixing appropriate amounts of dry (0% RH) and wet (100% RH) nitrogen gas. The 3D stacked nanofibers were placed in the gas chamber of the flow system and exposed to controlled levels of formaldehyde gas (400 mL min⁻¹). For all sensing experiments, image acquisition was performed through an optical glass using a flatbed scanner (Perfection V550 Photo, Epson, Nagano, Japan). The RGB values and differential color maps were obtained by the aforementioned method.

**Characterization:** $^{1}$H NMR spectra were acquired using an NMR spectrometer (AVANCE III 300, Bruker, Massachusetts, United States) at 300 MHz. The $^{1}$H NMR samples included the 0.01 wt% ion-pairing dye solution in MeOH-d₄, DMF-d₇, and DMSO-d₆, for CV–EO, BG–MR, and BG–NO, respectively. The morphology of the prepared nanofibers was observed by SEM using a scanning electron microscope (JSM-6360LV, JEOL, Tokyo, Japan) at an acceleration voltage of 5 kV. The IR spectra were obtained using an FTIR spectrometer (Nicolet 6700, Thermo Fisher Scientific, Massachusetts, United States) at a resolution of 4 cm⁻¹ and with 64 scans. Spectroscopic analysis was performed using a UV–Vis spectrophotometer (LAMBERTA 360, Perkin Elmer, Massachusetts, United States) in the absorption mode in the wavelength range of 350–800 nm. The samples for the UV–Vis absorbance analysis were prepared by dropping 20 μL of the 0.1 wt% ion-pairing dye solution in MeOH into 3 mL of the buffer solution with various pH levels. The porosity and pore distribution were analyzed using a mercury intrusion porosimeter (Auto Pore V9600, Micromeritics, Georgia, United States) for details in Table S5, Supporting Information.

**Design and Evaluation of the Optoelectronic Filter System:** The filter system comprised three parts: a custom-designed cylindrical filter case, head dummy (My-51, Myungmoon Medical, Seoul, Republic of Korea), and gas mask with an LED (750X 7502, 3M, Minnesota, United States). The cylindrical filter case consisted of two RGB sensors (TCS3200, SMG, Guangdong, China) and provided a long flow path to facilitate the gas exposure of fillings (Figure S16, Supporting Information). The device operation and digital signal processing were controlled by a single-board microcontroller (UNO R3 SMD, SMG, Guangdong, China). Additionally, individual RGB sensors were calibrated by normalizing the data of DEW380 (warm white) and DEA187 (black) color charts (Perfect palette, Dunn-Edwards Paints, California, United States) for the maximum and minimum RGB values, respectively. These standards were meant for the one-time calibration of RGB sensors and hence, were employed for all experiments with this filter system. The analytical gas stream was generated by mixing humidified nitrogen and formaldehyde gases; the formaldehyde concentration was measured using the portable air quality monitor with a typical concentration and humidity of 20 ppm and 50% RH, respectively. An integrated development environment software (Arduino IDE 1.8.19, Arduino) for the RGB sensors was used to acquire the RGB values of the colorimetric membrane at a flow rate of 1000 mL min⁻¹.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

The data that support the findings of this study are available from the corresponding author upon reasonable request. Source data are provided with this article.

**Keywords**

artificial sensing systems, chemoresponsive dyes, colorimetric sensors, electrospinning, gas masks, ion-pairing formation, sensing arrays

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