

# Monitoring of Sweat pH and Dual-Mode Anti-Counterfeiting from Metal–Organic Framework-Based Multifunctional Gel

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## Abstract

Monitoring of sweat pH play important roles in physiological health, nutritional balance, psychological stress, and sports performance. The combination of functional MOFs with phosphorescent material to acquire the real-time physiological information, as well as the application of dual mode anti-counterfeiting, have seldom been reported. Herein, we reported MOF and phosphorescent dyes based multifunctional gel films with H<sup>+</sup> response and the related mechanism was studied in detail. Upon induction of H<sup>+</sup>, the composite gel film exhibited decreased fluorescent signal but enhanced room temperature phosphorescence (RTP), which could be utilized for sweat pH sensing through a dual-mode. Moreover, multifunctional gel films exhibited a potential application in information encryption and anti-counterfeiting by designing of stimulus responsive multiple patterns. This research opens a new avenue for portable and non-invasive sweat pH monitoring method, provides opportunities for effective anti-counterfeiting, also offers new insights stimulus-responsive multifunctional materials and their potential applications.

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Monitoring of Sweat pH and Dual-Mode Anti-Counterfeiting from Metal–Organic Framework-Based Multifunctional Gel

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## Keywords

Metal–Organic Framework | Room-temperature phosphorescence | Fluorescence | Sweat pH monitor | Anti-counterfeiting  
**Comprehensive Summary**

Monitoring of sweat pH play important roles in physiological health, nutritional balance, psychological stress, and sports performance

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## Background and Originality Content

Sweat not only removes heat but also helps excrete other chemicals and metabolites from the body. The

lactate concentration of sweat, mainly contributes to sweat pH, depends on metabolism and physical activity, which links to energy metabolism for assessing fatigue. Besides, it can also be a potential biomarker for pressure ischemia, which leads to a shortage of oxygen that is needed for cellular metabolism.<sup>[1]</sup> Sweat pH beyond the usual range (4-7) may indicate various health issues,<sup>[2]</sup> thus, monitoring of sweat pH plays important roles in physiological health, nutritional balance, psychological stress, and sports performance.<sup>[3-7]</sup> Thus far, various non-invasive point-of-care-testing sensing systems based on enzymes have been developed, however, some disadvantages including high cost, poor stability and susceptibilities to environmental influences limit their practical applications.<sup>[8]</sup> Metal-organic frameworks (MOFs) are potential candidates for non-enzymatic biosensing due to their unique porous structure, high stability and tailored functionalities.<sup>[9-13]</sup> The integration of functional MOFs with wearable sensors provides important insights into noninvasive technologies to acquire real-time physiological information.<sup>[14-18]</sup> However, the direct monitoring of sweat pH based on MOF fluorescence sensor has seldom been reported.

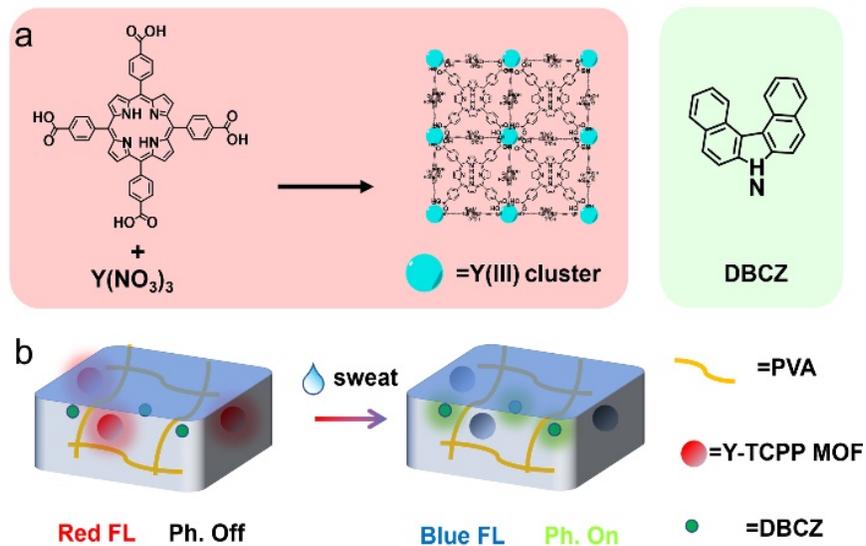
Recently, there has been a remarkable increase in the rational design of biosensor based on time-resolved emission characteristics. RTP with persistent luminescence, are more desired, owing to the disadvantages of the bare fluorescence probes including photo-bleaching, fluctuation of the excitation power and so on.<sup>[19-21]</sup> It was more desired that the fluorescent and phosphorescent dyes exhibited different emission wavelength and opposite stimuli-responsive behaviour, thus the relative changes in fluorescent and phosphorescent intensity might be utilized to monitor the sweat pH. Moreover, the combination of fluorescent and phosphorescent materials to construct dual-mode response provides new opportunities for anti-counterfeiting technology and practical application.<sup>[22-26]</sup> In this regard, it still remains a challenge to develop MOF-based functional composite materials with efficient dual-mode fluorescent and phosphorescent emission for sweat pH sensing and information encryption.

Herein, we prepared multifunctional composite gel films by the combination of fluorescent MOF and phosphorescent dyes Upon induction of  $H^+$ , the composite films exhibited decreased fluorescent signal but enhanced RTP, which could be utilized for sweat pH sensing through a dual-mode. In addition, the above multifunctional composite gel films with pH-responsive fluorescence and RTP exhibited a potential in information encryption and anti-counterfeiting application. This work not only provides in-depth understanding of the stimulus-responsive mechanism of MOF-based functional materials but also advances their potential applications toward sweat pH sensing and anti-counterfeiting.

## Results and Discussion

Firstly, Y-TCPP MOF was synthesized according to the previous report,<sup>[27]</sup> as shown in Figure 1a. The successful synthesis of Y-TCPP MOF was proved by scanning electron microscopy-X-ray energy dispersion spectroscopy (SEM-EDS, Figure S1), X-ray photoelectron spectroscopy (XPS, Figure S2) and X-ray diffraction (XRD, Figure S3). According the SEM-EDS and XPS spectra, it is substantiated the existence of C, N, O and Y. The characteristic peaks of 158.2 eV, 284.8 eV, 400.0 eV and 532.0 eV in XPS spectrum are attributed to the Y3d, C1s, N1s and O1s peaks of Y-TCPP MOF. In addition, the prepared Y-TCPP MOF powder showed multiple strong diffraction peaks at 5.82, 7.46 and 8.88, respectively, indicating the formation of MOF ordered structure. Moreover, the disk structure of Y-TCPP MOF can be clearly seen through SEM images, and the size of multiple disk structures stacked together is about 2  $\mu\text{m}$ . Based on the above results, Y-TCPP MOF which is consisted by porphyrin and Y clusters can be considered to be successfully synthesized. Unlike MOF of non-transition metals, Y in this material has more coordination sites to participate in the interaction.

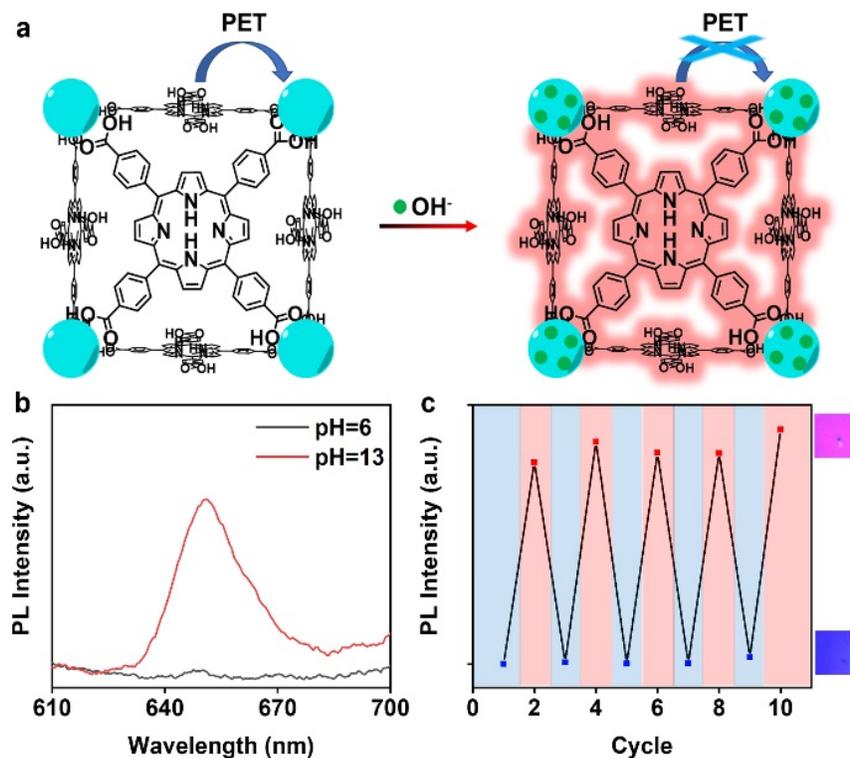
Y-TCPP MOF could emit the red fluorescence under 365 nm irradiation under alkaline condition<sup>[27]</sup>, while have no photoluminescent properties in the pH range of sweat. On the other hand, 7H-dibenzo[*c, g*]carbazole (DBCZ)<sup>[28]</sup> emits blue fluorescence and yellow-green RTP under 365 nm excitation.



The combination of the two luminous substances to prepare a multifunctional gel film brings the unexpected pH response properties in the pH range of sweat. Weakly acidic sweat can induce a decrease in the red fluorescence of MOF in the composite gel film, forcing the blue fluorescence of BDCZ to highlight, resulting in a red-blue transformation of the functional gel film visible to the naked eye. Besides sweat can promote the phosphorescence of DBCZ. Based on these, a dual-mode sweat pH sensor and sweat-based anti-counterfeiting can be designed (Figure 1b).

#### Properties of Y-TCPP/DBCZ PVA composite film

According to the previous reports, the fluorescence intensity of Y-TCPP MOF increases with the increase of pH from 6-12.<sup>[27]</sup> Although these properties are still maintained in the composite film, there have no responsive photoluminescent property in the pH range of sweat. Actually, this excellent alkali stimulation responsive luminescence performance of MOF provides an excellent platform for our composite luminescent materials.

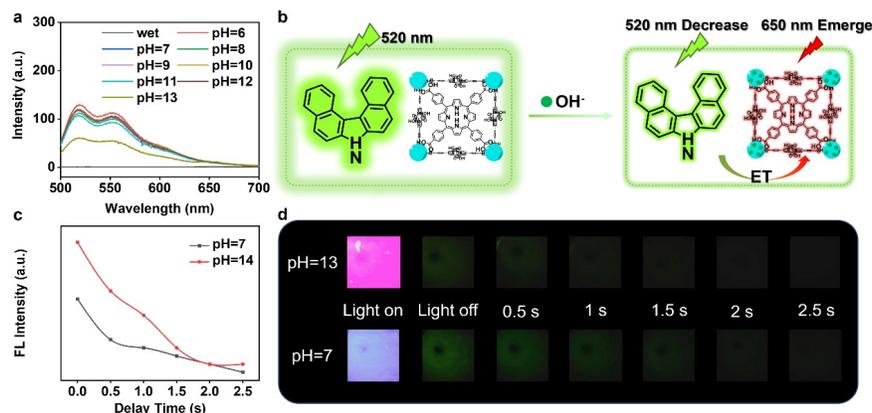


Fortunately, the multifunctional gel has been successfully prepared by compositing the PVA with MOF and DBCZ. We have screened various concentrations of MOF and DBCZ, 10 mg MOF and 2 mg DBCZ per 1 g PVA was found to be appropriate to ensure strong fluorescence intensity as well as obvious visually blue fluorescence under neutral conditions and red fluorescence under alkaline conditions. The composite film is constructed using the drop film method, and the required composite gel film can be obtained by evenly coating and drying. The presence of DBCZ and porphyrins can be observed through UV-Vis absorption spectroscopy (Figure S4). It is worth noting that PVA is specially treated. Commercially available PVA was highly susceptible to water erosion without exception although we have attempted many times, which undoubtedly hinders the construction of composite materials. Fortunately, choosing the method of glutaraldehyde crosslinking to improve the water erosion resistance of PVA could solve this problem. At the same time, considering the crosslinking efficiency and the influence of PVA embrittlement resulting from the concentration is too high, the concentration of glutaraldehyde in the crosslinking solution is 1.25%. It is confirmed that the film was completely immersed in the cross-linking solution for 5 min, there was no obvious water erosion on the gel film. There is no obvious change of surface morphology before and after treatment through microscope photos (Figure S5). In order to further characterize the success of crosslinking, the Fourier transform infrared spectroscopy was carried out before and after crosslinking. The C-O-C stretching vibration at  $1142\text{ cm}^{-1}$  is clearly visible, demonstrating the formation of acetals (Figure S6). Based on above, stimulus responsive films with good water resistance and specific response ability have been successfully synthesized.

#### Fluorescence behavior of Y-TCPP/DBCZ PVA gel film at different pH values

The luminous properties of composite material are the key to the design of multifunctional film. The emission of Y-TCPP/DBCZ PVA gel film with pH changes was recorded under 365 nm excitation. The fluorescence of DBCZ remains basically unchanged at pH greater than 6 (Figure S7), while the fluorescence intensity of MOF increases with the increase of OH<sup>-</sup> concentration as previously reported (Figure S8). The mechanism of fluorescence enhancement of MOF is attributed to the photo induced electron transfer (PET)(Figure 2a). It is

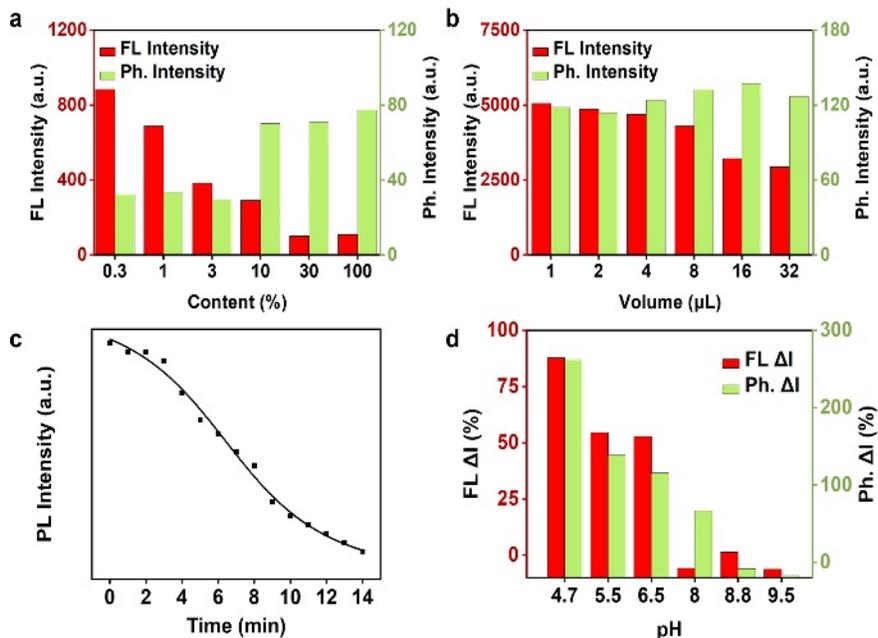
worth noting that the MOF emission is obviously change until the pH up to 13 (Figure S9). The fluorescence photos are corresponding well to the results of spectroscopy. The blue fluorescence was observed at pH=6-12, and tune to the red when pH become 13, which may be due to that the weak fluorescence intensity of MOF at pH of 6-12 and the blue fluorescence emission of DBCZ is dominant. When pH=13, prohibited PET results in the red fluorescence emission of the film (Figure 2b) thus red is more visible to the naked eye. All above experimental results indicate that there is a mutation in the fluorescence color of the composite film from pH=12 to pH=13.



In order to further improve the application value of the composite material, the repeatability of the luminescence intensity caused by pH is necessary. However, immersing a red film with pH=13 into a Tris solution with pH=6 did not restore the blue color. We believe that this may be due to the excessive amount of OH<sup>-</sup> adsorbed in the film and is not completely neutralized. For this reason, the idea of using acids with a lower pH to neutralize excessive alkalis was proposed. However, the acetal structure will undergo hydrolysis in the acid, which clearly damages the film itself. Fortunately, the crosslinking solution itself is strongly acidic and can be happens to neutralize alkali. Putting a red film with pH=13 into the crosslinking solution, the film quickly turned blue in less than a minute, and the morphology of the film did not change significantly. Besides, in the repeatability experiment characterized by fluorescence intensity, the fluorescence intensity of gel film showed good repeatability in pH modulation of pH=13 solution and cross-linking solution (Figure 2c).

#### Phosphorescence behavior of Y-TCPP/DBCZ PVA mixed gel at different pH values

RTP has a wide range of applications in multi-field. In order to investigate the phosphorescence property of the composite films, the effect of pH value on the yellow-green phosphorescence of DBCZ-MOF PVA composite films were detailed studied. Phosphorescence can hardly be observed in wet films, which is consistent with reports that water can quench phosphorescence.<sup>[29]</sup> In this case, the films were immersed in Tris solution at pH=6 to 13 for 10 minutes and well dried it at 40 °C subsequently to be tested. As expected, there was no significant change in the phosphorescence of the dried film from pH=6 to pH=12, and exhibited a jump variation in phosphorescence between pH=12 and 13, which is similar to the phenomenon discussed before. When the pH increased to 13, the phosphorescence intensity of the film decreased significantly, and a porphyrin emission peak (650 nm) appeared, which is generally considered delayed fluorescence (Figure 3a). This phenomenon is attributed to energy transfer (Figure 3b).



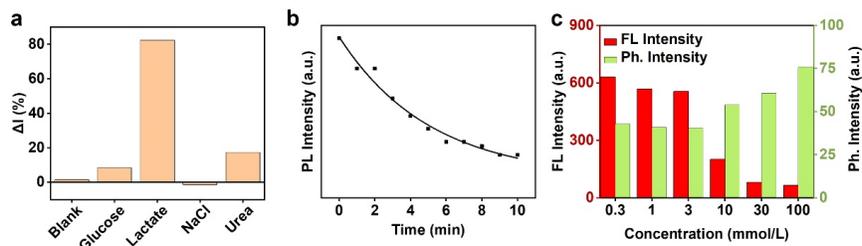
The changes in phosphorescence can be clearly observed in the macroscopic photos of the composite film under different states, and the emission intensity and luminescence lifetime of the film with pH=13 show a significant decrease compared to the film with pH=7 (Figure 3c). Actually, phosphorescence behavior of Y-TCPP/DBCZ PVA mixed gel at different pH values were studied in detail (Figure 3d). The composite film exhibits blue fluorescence and yellow-green RTP at pH=6 to pH=12. When the pH rises to 13, the fluorescence of the film turns red, and the RTP intensity significantly decreases, accompanied by a decrease in luminescence lifetime.

### Monitoring of Sweat pH

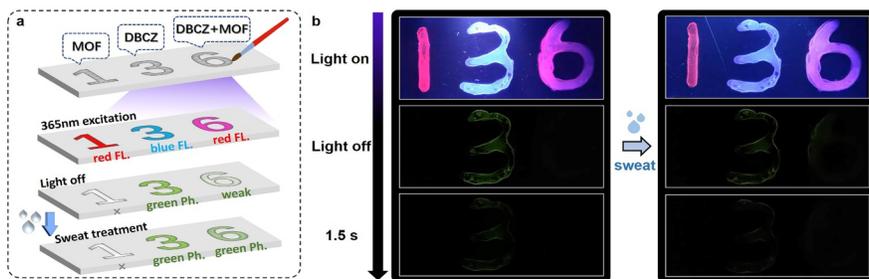
In human bodily fluids, sweat is a weakly acidic liquid (4-7) with a wider pH range compared to other bodily fluids, such as blood. Sweat pH sensor can be used to diagnose metabolic alkalosis, which is a metabolic disease. When the pH value of tissue rises beyond the normal range, it will lead to various health problems, such as arrhythmia, abnormal sensation, neuromuscular irritability, etc., and even Seizure and coma in severe cases. Inspired by the photoluminescence behavior of the pH responsive of composite film, a method for monitoring of sweat pH is proposed here.

The pH of sweat is mainly determined by the concentration of lactic acid. The discussion about the effect of lactic acid on the luminescence of the composite film is necessary. Considering the weak acidity of sweat itself and the luminescence change of gel film under alkaline conditions, the gel film was soaked in Tris solution with pH=13 and then dried it. Four common substances in sweat, lactic acid, glucose, NaCl, and urea, were selected to investigate the response selectivity of the gel film. 50 µL 10 mmol·L<sup>-1</sup> solution of selected substance was applied to the film and dried. Compared with the original state, lactic acid treatment showed the most significant fluorescence attenuation, while urea had little effect. The effects of glucose and NaCl on the fluorescence of the composite film were almost invisible (Figure 4a). These results demonstrate the selective response towards lactic acid for the composite film. In order to investigate the effect of lactic acid on the luminescence of the gel films, further research is needed on response time and detection limit. Adding 10 µL lactic acid solution of 10 mmol·L<sup>-1</sup> onto the gel film and then recording the fluorescence intensity. It is obviously that the fluorescence intensity remains basically unchanged after 6 minutes (Figure 4b). From the graph of the relationship of lactate concentration and the luminescence intensity of the gel film, the fluorescence intensity decreases significantly after the lactate concentration exceeds 3 mmol, while the

phosphorescence intensity increases significantly (Figure 4c). The above results indicate that the composite gel film can be applied to the detection of lactic acid in terms of fluorescent changes.



On the basis of lactic acid detection discussed above, a sweat pH sensor based on Y-TCPP/DBCZ PVA composite gel film was designed, as shown in Figure 5. In order to ensure the repeatability of the experiment, artificial sweat was selected as the test sample to avoid the difference in pH. Artificial sweat with pH=4.7 was selected as the base sample. 50  $\mu$ L different proportions of sweat were applied to the film and dried, it is found that when the proportion of sweat was higher than 3%, the intensity fluorescence and phosphorescence have changed dramatically (Figure 5a). We also investigated the effect of coating amount on the luminescence of the composite gel films, the higher of the amount of the sweat applied at, the lower of the fluorescence intensity, and the stronger of the phosphorescence intensity. However, due to the different sizes and shapes of sweat stains after drying, the differences are not obvious (Figure 5b). The response time to sweat is also a topic that needs to be explored, and we found that the 10  $\mu$ L sweat droplets on the film show little change in the first four minutes, also remain basically unchanged after 12 minutes (Figure 5c). Finally, the influence of artificial sweat with different pH on the luminescence of coincidence membranes was explored in order to expand the application of coincidence membranes in health diagnosis. The higher the pH value, the smaller the change in luminescence intensity (Figure 5d). From the above results, a sweat pH sensor has successfully constructed based on Y-TCPP/DBCZ PVA composite gel film. This composite material has application prospects in body fluid detection and health examination.



### Dual-mode Anti-counterfeiting

With the development of information technology and economy, the demand for anti-counterfeiting technology is becoming increasingly urgent in many fields such as food, medicine, clothing, and communication. The most common single color anti-counterfeiting is limited due to its single mode practical application. Herein we present a multimode anti-counterfeiting film based on Y-TCPP/DBCZ PVA composite gel film (Figure 6a). Considering the risk of MOF being damaged by direct exposure to alkali, writing with PVA dispersion of DBCZ and MOF is more suitable than applying MOF and DBCZ onto PVA films. We prepared multifunctional gel 1 (a dispersion of 1 mg MOF, 100 mg PVA, and 4 mL water), gel 2 (a dispersion of 0.2 mg DBCZ, 100 mg PVA, and 4 mL water), and gel 3 (a dispersion of 1 mg MOF, 0.2 mg DBCZ, 100 mg PVA, and 4 mL water) as ink for writing. Afterwards, use multifunctional gel 1, 2, and 3 to write the numbers "1", "3", and "6" on the substance, respectively. Due to insufficient sample size to construct continuous

strokes, additional multifunctional gel is added to the corresponding stroke. After drying and cross-linking, the composite gel film is immersed in a Tris solution with pH=13. After the fluorescence turns red, excess alkaline solution is wiped off and dried. The dried film exhibits obvious red "1", blue "3", and rose red "6" under excitation of 365 nm light. After turning off ultraviolet light, a yellow-green "3" and a faintly visible "6" can be seen. After soaking the film in an artificial sweat solution at pH=4.7 for 20 minutes and drying, the light red "1", blue "3", and light purple "6" were displayed under excitation of 365 nm light. After turning off ultraviolet light, the yellow-green "3" remains unchanged from before sweat treatment, and the weaker lightness "6" is clearly visible (Figure 6b). These experimental results prove the application prospect of Y-TCPP/DBCZ PVA mixed gel in dual-mode anti-counterfeiting materials, and show the application value of organic afterglow materials with adjustable solid emission in anti-counterfeiting.

## Conclusions

Based on the advantage of Y-TCPP MOF for OH<sup>-</sup> specific response, a two-color stimulus response gel was constructed in combination with DBCZ PVA system, and a sweat pH sensing and anti-counterfeiting mode that is economical, applicable, easy to prepare and operate was found. A MOF with specific response to OH<sup>-</sup> was synthesized and combined with the DBCZ PVA system to construct a stimulus responsive thin film material with fluorescence red blue color changing with pH, which has good repeatability. The phosphorescence of the mixed system exhibits a significant decrease in emission intensity and luminescence lifetime at pH=13. The above spectroscopy results are visible to the naked eye. A sweat pH sensor was constructed and exhibited different luminescence changes under different conditions. The effect of different pH sweat on luminescence may be used for health testing. A fluorescent phosphorescent composite anti-counterfeiting material has been constructed, which can achieve dual-mode anti-counterfeiting performance. Under alkaline and sweat conditions, it exhibits different fluorescence and phosphorescence properties visible to the naked eye.

## Experimental

### Materials

Yttrium nitrate hexahydrate (Y(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O, 99.9%), 7H-dibenzo[c, g]carbazole (DBCZ, 98%), polyvinyl alcohol 1799 (PVA, 1799 type, alcoholysis degree 98-99%), lactate (90%) were purchased from Aladdin Co., Ltd. Tetra-(4-carboxyphenyl) porphyrin (TCPP, 95%) was purchased from Leyan Co., Ltd. Glutaraldehyde (25% aqueous solution), hydrochloric acid (36%-38% HCl aqueous solution), urea (99.0%), glucose (AR), tris-(hydroxymethyl)aminomethane (Tris, 99.5%), sodium hydroxide (NaOH, 99%) sodium chloride (NaCl, 99.5%) were purchased from Sinopharm Chemical Reagent Co., Ltd. Artificial sweat were purchased from Shenzhen Zhongwei equipment co., LTD. All other solvents and reagents were of analytical grade and used as received. Milli-Q water (18.2MΩ·cm) was used in all cases.

### Instruments

Use GeminiSEM 500 to record the scanning electron microscope (SEM) image and the X-ray energy dispersion spectrum (EDS) image. The steady-state fluorescence and phosphorescence measurements were obtained on the Hitachi F-4600 fluorescence spectrophotometer at room temperature. X-ray photoelectron spectroscopy (XPS) was performed on ESCALAB 250Xi X-ray photoelectron spectrometer. X-ray diffraction (XRD) was performed on the Rigaku SmartLab 9kW. Use SHIMADZU UV-2700 PC spectrophotometer to measure UV-vis absorption spectrum. Fourier transform infrared spectroscopy (FT-IR) was measured by using BRUKER TENSOR II.

### Preparation of Y-TCPP MOF

The synthesis method is taken from the previous report.<sup>[27]</sup> TCPP (21.0 mg, 0.0266 mmol) was completely dissolved in DMF (45.0 mL) in a 100 mL round bottom flask. Y(NO<sub>3</sub>)<sub>3</sub>·6H<sub>2</sub>O (15.0 mg, 0.0392 mmol) was dissolved in 3 mL ultra-pure water. Then Y(NO<sub>3</sub>)<sub>3</sub> solution was added into TCPP solution and stirred evenly. The mixture was stirred and heated to 60 °C for 2 h, further heated to 100 °C for 2 h. After cooling, the reacted liquid was centrifuged at 10000 rpm for 25 min, collected the precipitation, and washed twice with

ethanol to obtain brown solid. The brown product attached to the bottle wall could be dispersed by soaking in anhydrous ethanol. Finally, Y-TCPP was stored in ethanol and need to be shaken to uniform dispersion before use. The concentration of the dispersion solution is obtained by measuring a quantitative liquid in a bottle and weighing the solid after drying in a vacuum oven at 60 °C for 30 hours.

#### Preparation of Y-TCPP/DBCZ PVA mixed gel

100 mg of PVA was added into a glass bottle containing 4 mL of ultra-pure water, and heated at 90 °C until it is completely dissolved. After cooling, 1 mg of centrifuged Y-TCPP MOF was added and the mixed solution was ultrasonicated until well-distributed. 1 mg of DBCZ was dissolved in 0.1 mL of ethanol and ultrasonicated until it is fully dissolved. 20  $\mu$ L DBCZ solution was added to PVA solution and stirred for 0.5 h. The dispersion solution is light brown and unobvious turbidity was observed. 0.1 mL of sample was dropped onto a glass sheet and dried in air at 40 °C for 1 hour to form the gel film.

#### Preparation of water-resistant mixed gel

0.15 mL of 25% glutaraldehyde, 0.25 mL of concentrated hydrochloric acid and 2.6 mL of acetone was stirring evenly as the cross-linking solution. The gel film was completely immersed in a 5 mL beaker containing 3 mL cross-linking solution for 5 min, then take it out and blow off the excess liquid with nitrogen, placed in a ventilated environment for 1 h. The gel film is stable and can not be eroded by water.

#### Supporting Information

The supporting information for this article is available on the WWW under <https://doi.org/10.1002/cjoc.2023xxxxx>.

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## The Authors

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## Entry for the Table of Contents

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**Monitoring of Sweat pH and Dual-Mode Anti-Counterfeiting from Metal–Organic Framework-Based Multifunctional Gels**  
The MOF/DBCZ-based multifunctional gel with pH triggered fluorescence and RTP variation was applied in monitoring of

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