



## Conversion of absorption to fluorescence probe in solid-state sensor for nitric oxide and nitrite



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### ARTICLE INFO

#### Article history:

Received 17 September 2012

Received in revised form 30 November 2012

Accepted 11 December 2012

Available online 5 January 2013

#### Keywords:

Nitric oxide

Solid-state real-time sensor

1,2-Diaminoanthraquinone

Fluorescence

### ABSTRACT

A hydrogel bi-layer structure is used to convert an absorption type to a more convenient fluorescent type probe film for aqueous nitric oxide (NO) and nitrite (NO<sub>2</sub><sup>-</sup>). In the bottom layer the commercial probe molecule 1,2-diaminoanthraquinone (DAQ) is dispersed in poly 2-hydroxyethyl methacrylate (poly HEMA), in the top layer the red inorganic phosphor is dispersed in poly HEMA. Both optical excitation and detection is from the bottom. This bi-layer is integrated with organic light-emitting diode (OLED) and organic photo-detector to form a real-time solid-state sensor. The sensor is responsive to NO bubbling, and remains stable in acid condition.

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### 1. Introduction

Nitric oxide (NO) is a key biochemical messenger (a biological signaling molecule) which plays important roles in blood pressure regulation and immune systems [1–6]. NO has a short lifetime in body fluid and decay into nitrite (NO<sub>2</sub><sup>-</sup>) in the presence of oxygen [7–13]. Nitrite is an oxidative breakdown product of NO synthesis and has important biological functions including maintenance of vascular homeostasis as well [14–17]. A solid-state real-time sensor for nitric oxide and nitrite will be an important device for related biomedical research and diagnosis. The key component of such sensor is a probe material whose optical properties are modulated by the target molecules with high specificity. 1,2-Diaminoanthraquinone (DAQ) is so far the only commercial nitric oxide and nitrite probe molecules

with high specificity and low cost. Unfortunately it is an absorption type probe rather than the common fluorescence type probe [7,18–24]. In the common fluorescence type probe the light emission is turned on, or turned off, only when particular chemical reaction with the target molecules takes place. The detection wavelength  $\lambda_{pl}$  is usually away from the excitation wavelength  $\lambda_{ex}$ . On the other hand in the absorption type probe the detection and excitation can only be done at the same wavelength  $\lambda_{ex}$  as there is no emission at a longer wavelength. The problem of absorption type probe for solid-state device is that the excitation light intensity can be disturbed by various other factors including change in light scattering and refractive index. It is therefore difficult to separate the small real signal of the aimed chemical reaction from the other factors. Apparently there is no such mixing of signals in fluorescence type probe as the emission signal exclusively results from the aimed reaction, and is not undermined by the small fluctuation of the optical excitation. Furthermore, in the integrated opto-electronic sensor both the light

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source and photo-detector are on the same side of the sensing film which is in contact with the fluid [25–31]. For absorption type film the light source and the photo-detector have to be separated by the sensing film and the fluid, the device structure is therefore complex and inconvenient to use.

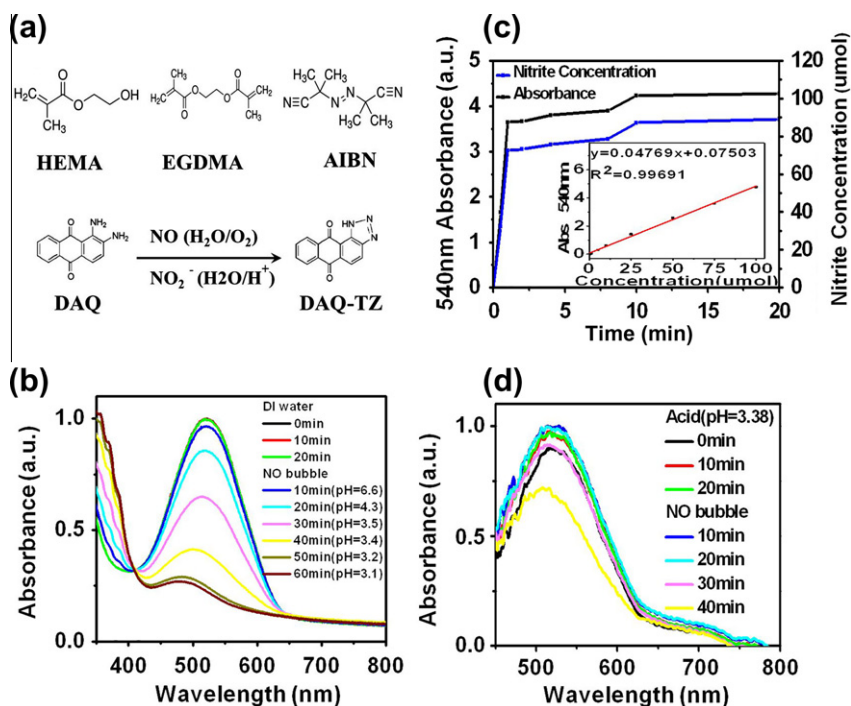
Furthermore, in case that the fluid is between the excitation source and photo-detector, any variation in the optical properties of the fluid in between will also affect the photo-current in addition to the real sensing film signal. With the conversion film the photo-detector and excitation are on the same side, so the variation of the optical properties of the fluid will not affect the signal.

In this work we present a bi-layer structure which converts the inconvenient absorption type sensing film into a fluorescence sensing film based on DAQ. The bottom layer contains DAQ molecules dispersed in a hydrogel poly 2-hydroxyethyl methacrylate (poly HEMA) with good water permeability, and the top layer contains an inorganic phosphor also dispersed in the hydrogel. When the bi-layer is immersed in fluid the nitric oxide and nitrite molecules will diffuse through the hydrogel and change the absorption spectrum of the bottom film. The excitation light comes from bottom and passes through DAQ film. The phosphor fluorescence is proportional to the light intensity after passing through the bottom layer and reflects the modulation of the absorption of DAQ. In this way the phosphor emission at  $\lambda_{PL}$  can be easily separated from the excitation  $\lambda_{ex}$  tuned at DAQ absorption peak, and the fluorescence can be conveniently collected by the photo-

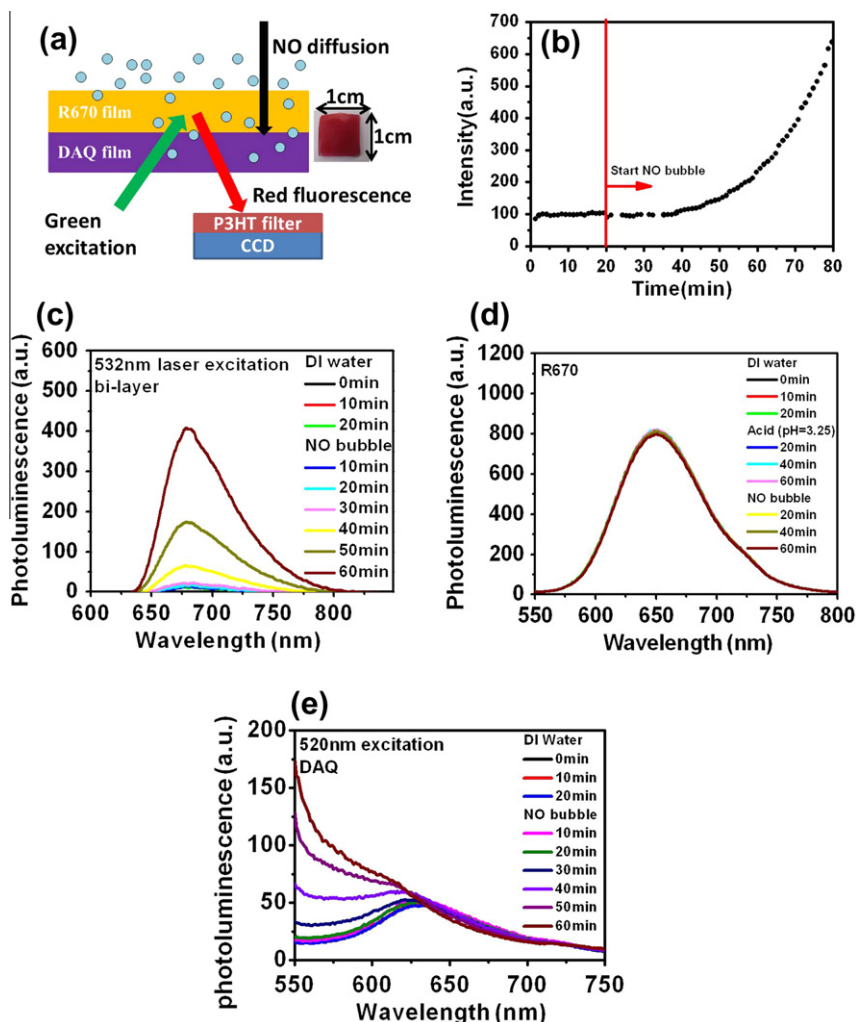
detector underneath the bi-layer as the case of common fluorescence type film [26,27]. Under NO bubbling in deionized water (DI water) the red fluorescence peaked at 670 nm from the phosphor increases in the bi-layer structure, because the absorption of the green excitation around 520 nm by the DAQ bottom layer is reduced. The bi-layer structure is shown to be stable in acid condition so the possibility of signal due to pH variation is easily ruled out, in contrast to the previous report of fluorescence probe which suffers from instability under acid condition [25,26]. Integrated device is made by using organic light-emitting diode (OLED) as the green light source. Both Si photo-detector and an organic photo-detector with a low band-gap polymer are used to collect the red emission from the phosphor in real time. A solid-state integrated fluorescence type sensor for nitric oxide and nitrite is therefore formed by a commercial absorption type probe DAQ.

## 2. Experimental

The hydrogel film is made by mixing the monomer 2-Hydroxyethyl methacrylate (HEMA), the cross-linker Ethylene glycol dimethacrylate (EGDMA), and the thermal initiator Azobisisobutyronitrile (AIBN) shown in Fig. 1a. The weight ratio between HEMA and EGDMA is 12:1, the weight concentration of AIBN is 1 wt.%. 1,2-Diaminoanthraquinone (DAQ) is dissolved in the liquid mixture of the three with weight concentration of 0.06 wt.%. The solution with volume 0.8 ml is annealed at 80 °C for 12 min in a



**Fig. 1.** (a) The chemical structures of the HEMA, EDGMA, AIBN, and DAQ. The reaction of the DAQ. (b) The absorbance of poly HEMA:DAQ with 0.06 wt.% is shown for various NO bubbling time. The green absorption band around 520 nm decreases with time. The pH versus time is also shown. (c) The absorbance at 540 nm is plotted against calibrated nitrite concentration. Using this relation the nitrite concentration at various NO bubbling time is shown. (d) The absorbance of the poly HEMA:DAQ film is shown at acid condition of pH = 3.38. Before NO bubbling the absorbance does not change, implying its stability.



**Fig. 2.** (a) The bi-layer structure to convert the absorbance modulation of DAQ into the fluorescence signal of the R670 phosphor is shown. Both DAQ and R670 are dispersed in poly HEMA. Green optical excitation from the bottom is able to reach R670 only after passing through the absorbing DAQ film. The red fluorescence from R670 is collected from the bottom. The bi-layer sensing hydrogel film is also shown. (b) The R670 fluorescence signal taken every minute is shown. (c) The R670 fluorescence of the top film increases with NO bubbling time. The excitation is a 532 nm diode laser from the bottom. The PL spectrum is taken by a CCD covered by a P3HT filter. (d) The top poly HEMA:R670 film is shown to be stable against NO bubbling and acidity. (e) The red fluorescence of reacted DAQ is below the detection limit of the PL spectrometer. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

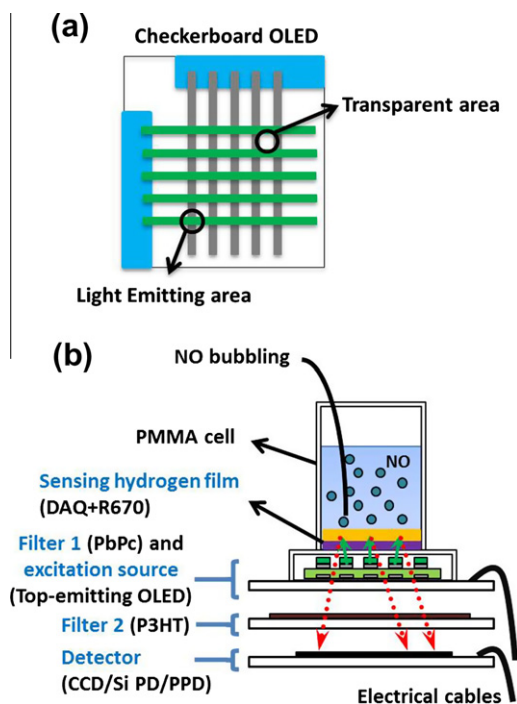
Teflon mold [7], resulting in a bottom sensing film with length 4.5 cm, width 3 cm, and thickness 0.6 mm. The top conversion film is made in the same way, and it is also 0.6 mm thick, except that DAQ is replaced by the red inorganic phosphor R670 purchased from Intematix. The weight concentration of R670 is 2 wt.%. The top conversion layer and bottom sensing layer are laminated and they attach to each other tightly only by Van der Waals forces. The chemical structures and reactions are shown in Fig. 1a.

Both the reaction of DAQ with NO and the decay of NO involve the dissolved oxygen molecules in deionized water (DI water) [7].

The emissive layer of the top-emitting green organic light-emitting diode (OLED) is a 65 nm film of the Green B conjugated polymer from Dow Chemical. The device structure is TFB/Green B/Ca (10 nm)/Ag (15 nm)/PbPc (100 nm). The Lead phthalocyanine (PbPc) top filter is

evaporated together with the top semi-transparent cathode with the same mask, its purpose is to remove the long wavelength part of the Green B emission. The details of OLED fabrication have been reported before [27]. The top-emitting OLED active area is made in a checkerboard pattern defined by the bottom Indium Tin Oxide (ITO) stripe and orthogonal top cathode stripes as shown in Fig. 3a. The size of each square is 1 mm by 1 mm in the checkerboard. Poly(3-hexylthiophene-2,5-diyl), regioregular (P3HT) filter of 250 nm on glass is used as the filter before the photo-detector, its purpose is to remove Green B emission from entering the photo-detector so only the R670 red emission is collected.

The organic photo-detector structure is PEDOT:PSS/PBDTTT-C-T:C71-PCBM/Ca (35 nm)/Al (100 nm). The 40 nm hole injection layer of poly-(3,4-ethylenedioxythiophene):poly-(styrenesulfonate) (PEDOT:PSS) is spin coated



**Fig. 3.** (a) The schematic diagram of the checkerboard top-emitting OLED (blue region: ITO electrode, gray region: anode and green region: cathode). (b) The integrated device structure is shown. A PMMA cell without bottom is glued on the top glass of the top-emitting OLED in checkerboard array. The sensing bi-layer film is placed on the top glass within the cell and immersed in DI water. NO is bubbled into the cell. The downward red fluorescence from R670 passed through the OLED checkerboard opening and collected by the photo-detector. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

on pre-cleaned ITO glass substrate and then annealed at 200 °C for 15 min. Poly[(4,8-bis[5-(2-ethylhexyl)thiophene-2-yl]benzo[1,2-b:4,5-b']dithiophene)-2,6-diyl-alt-(4-(2-ethylhexanoyl)-thieno[3,4-b]thiophene))-2,6-diyl] (PBDTTT-C-T) is the low band-gap polymer developed for organic solar cell [32]. [6,6]-Phenyl C<sub>71</sub> butyric acid methyl ester, mixture of isomers (C71-PCBM) is the electron acceptor. The active layer of PBDTTT-C-T:C71-PCBM(1:1.5, w/w, 3%DIO) is formed by blade coating on the PEDOT:PSS layer. The device is then completed by evaporating Ca/Al metal cathode.

For the real-time detection the bi-layer is placed on the bottom of a Polymethylmethacrylate (PMMA) cell as shown in Fig. 3b with 3 ml of DI water. 1% NO gas diluted by nitrogen is bubbled into the cell with the gas flow rate of 30 ml/min. In the integrated device the bottom of the PMMA cell is removed and the cell wall is fixed on the top encapsulation glass of the OLED by Ultraviolet (UV) epoxy as shown in Fig. 3b. The Si photo-detector is under -10 V reverse bias, and the organic photo-detector is under -5 V reverse bias.

### 3. Results and discussions

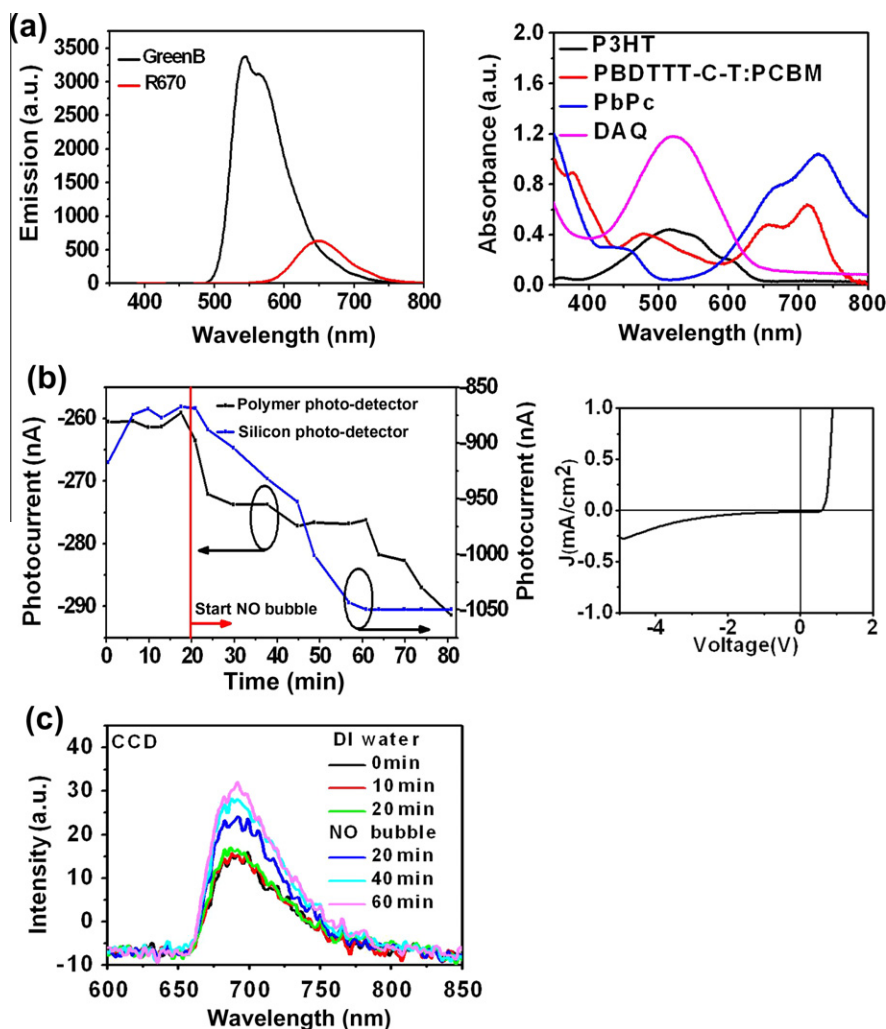
The bottom sensing film of DAQ dispersed in poly HEMA host responds to NO bubbling as shown in Fig. 1b. The

absorption peak around 520 nm is decreasing after 20 min, consistent with the previous report [7]. As discussed above nitrite NO<sub>2</sub><sup>-</sup> is one of the decay products of the short-lived NO in solution and its concentration accumulates with time. DAQ reacts with both NO and nitrite [7], the latter reaction dominates in the acid condition. pH value versus time is shown as inset in Fig. 1b. After 20 min of NO bubbling, the solution becomes acid so the decrease of 520 nm absorption is assumed to be primarily due to nitrite. The absolute concentration of nitrite can be obtained by the absorbance defined as  $-\log T$  where  $T = \exp(-\alpha d)$  is the transmission,  $\alpha$  is the 540 nm nitrite absorption coefficient and  $d$  is the optical path. For given nitrite concentration obtained by adding sodium nitrite in DI water, the calibrated absorbance-concentration relation is shown as inset in Fig. 1c. Using this relation the nitrite concentration is shown as a function of NO bubbling time in Fig. 1c. Before 10 min the solution is nearly neutral and NO is expected to be the primary reaction, but the reaction is weak and within the noise level. In order to rule out that the change in absorption in Fig. 1b is due to pH variation rather than NO/nitrite reaction, the sensing film is placed in an acid solution for 20 min without NO bubbling. The absorption does not change as shown in Fig. 1d, but started to decrease after NO bubbling in the acid solution. This result therefore proves that the signal is not due to the pH variation in Fig. 1b.

Next we turn to the bi-layer structure, shown in Fig. 2a, which converts the absorption to fluorescence signal. The bi-layer is excited by a 532 nm diode laser from the bottom as shown in Fig. 2a. After passing through the bottom sensing layer the 532 nm light reaches the top conversion layer. The intensity of the exciting light therefore depends on the absorbance of the green absorption band around 520 nm of the sensing film shown in Fig. 1b. During device operation both the entire bi-layer film in Fig. 2a is immersed in the fluid. The advantage of hydrogel host for the top layer is that NO and nitrite molecules can easily diffuse downward through the R670 sensing film to reach the bottom DAQ film, where the sensing chemical reaction actually takes place. Without the water permeability of the hydrogel film, NO and nitrite will be blocked by the conversion layer and can never reach the bottom sensing film. After reaction with NO or nitrite the green absorption band of DAQ decreases and the sensing film gradually becomes transparent. So the R670 red fluorescence of the top conversion film becomes stronger. The result is shown in Fig. 2c. P3HT filter is already applied to remove the spectral part shorter than 640 nm. Significant increase of the R670 emission after 20 min of NO bubbling implies that there is no barrier for NO or nitrite to penetrate the top conversion layer and enter the bottom sensing layer. The top conversion layer therefore converts the absorption to fluorescence signal without blocking the diffusion of the targeted chemicals. In this way the effective function of the bi-layer is the same as a single fluorescence sensing layer commonly used in the solid-state sensor for other biochemical molecules [27,33–36]. The inorganic phosphor R670 dispersed in poly HEMA is quite stable against pH variation and NO bubbling as shown in Fig. 2d. So all the signals in Fig. 2c is exclusively due to the modulation of the bottom layer by NO bubbling.

Finally the fluorescence signal from the top conversion layer is registered by a set of solid-state opto-electronic devices as shown in Fig. 3b. The green emission from the OLED is filtered by PbPc on top of the top semi-transparent cathode in order to remove the red tail. Without such PbPc filter the red tail would mix with the R670 fluorescence signal and becomes inseparable in the photo-detector output. The filtered green excitation passes through the sensing hydrogel film and excites the top conversion film. The downward red emission passes through the opening of the checkerboard shown in Fig. 3a and reaches the P3HT filter, which removes the green excitation and allows only the red emission from the conversion layer to reach the photo-detector in Fig. 3b. This red fluorescence can be readily registered by a Si photo-detector or organic photo-detector. The emission or absorption spectra of all the components are shown in Fig. 4a. The time-dependent photo-current of the Si photo-detector under NO bubbling

is shown in Fig. 4b. The red emission spectrum is checked by a CCD fiber spectrometer shown in Fig. 4c. Similar to the previous absorption results of the sensing film in Fig. 2c, after 20 min of bubbling a increase of the photo-current is seen in Si photo-detector. The NO or nitrite variation in solution is therefore detected by an all-solid-state integrated devices in real time. Organic light-emitting diodes and photo-detectors have the advantages over Si devices of easy fabrication on glass substrates. It is therefore possible to fabricate both the light source and photo-detector on the same chip at low cost. A low band-gap conjugated polymer PBDTTT-C-T is mixed with C71-PCBM to form the active layer of the organic photo-detector on glass substrate as discussed above. The results are shown in Fig. 4b with NO bubbling. Despite of the relatively large noise level the photo-current starts to increase also after about 20 min of bubbling time. The chemical signal is therefore registered by an all-organic integrated device.



**Fig. 4.** (a) The emission spectrum of the Green B OLED and R670 red phosphor are shown. DAQ absorption is also shown as reference. The absorption spectrum of the short-pass filter PbPc, the long-pass filter P3HT and the low band-gap polymer PBDTTT-C-T:PCBM are shown. (b) The dynamical responses of the Si photo-detector and organic photo-detector to NO bubbling are shown. The dark IV curve of the organic photo-detector is also shown (c) The spectrum after the P3HT filter and before the photo-detector is checked by a CCD are shown. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

#### 4. Conclusion

In conclusion, a desired fluorescent sensing film for key biochemical messenger NO and nitrite is realized using the low-cost absorption type probe molecule DAQ by a bi-layer hydrogel structure. When NO is bubbled in the solution, the bi-layer film converts the bottom absorption modulation into a top fluorescent signal which is easily collected. This bi-layer sensing film is integrated with organic LED and photo-detector to realize a solid-state sensor. The limit of detection is therefore  $10^{-4}$  M in the given device. This can be used as a low-cost biomedical device to detect the NO and nitrite in body fluid.

#### Acknowledgements

This work is supported by National Science Council of Taiwan under Contract Nos. 99-2628-M-009-002- and 98-2112-M-007-028-MY3, and the Veterans General Hospitals and University System of Taiwan Joint Research Program under Contract No. VGHUST100-G5-1-1, and the Academic Sinica under Contract No. AS-100-TP-B03.

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