Flexible, organic light-pen input device with integrated display

Seiichi Takamatsu\textsuperscript{a}, Maria Nikoloub\textsuperscript{b}, Daniel A. Bernards\textsuperscript{b}, John DeFranco\textsuperscript{b}, George G. Malliaras\textsuperscript{b,}\textsuperscript{∗}, Kiyoshi Matsumotoa, Isao Shimoyama\textsuperscript{a,}\textsuperscript{∗}

\textsuperscript{a} Department of Mechano-Informatics, The University of Tokyo, Bunkyo-ku, Tokyo, 113-8656, Japan
\textsuperscript{b} Department of Materials Science and Engineering, Cornell University, Ithaca, NY, 14853-1501, USA

\textbf{A R T I C L E I N F O}

\textbf{Article history:}
Received 4 October 2007
Received in revised form 10 June 2008
Accepted 5 August 2008
Available online 15 August 2008

\textbf{Keywords:}
Display
Photosensor
BacterioRhodopsin
Conducting polymer
PEDOT:PSS

\textbf{A B S T R A C T}

We report a light-pen input device with an integrated electrochromic display. The former is a photosensor array, based on a blend of bacterioRhodopsin (bR) and the conducting polymer poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), while the latter is based on PEDOT:PSS. The use of organic materials allows the entire device to be deposited on a thin, mechanically flexible plastic substrate. The operation and characteristics of this device are discussed.

© 2008 Elsevier B.V. All rights reserved.

\textbf{1. Introduction}

Human interface devices, such as keyboards and displays, play a major role in information exchange between human and computers. Therefore, there is an ever-present demand for new types of interface devices with improved usability [1,2]. Of particular interest are devices that integrate a display with an input component such as a touch-pad, as they allow input of information in an intuitive, straightforward manner [3]. In recent years, the display technology has advanced dramatically, and mechanically flexible displays based on organic semiconductors and on electronic-paper have been demonstrated [4,5]. However, the flexibility of the display causes several problems for the integration of input devices, as the latter also need to be flexible and to work when bent.

The most common input device is the touch-pad. It consists of two transparent electrodes separated by an air gap. Pressure in these devices results in a change of the capacitance or resistance between electrodes, allowing an easy measurement of the pressure profile [6]. However, upon bending, the dimensions of the air gaps change and the sensitivity across such a touch-pad varies, making the integration with flexible displays challenging. Therefore, an input device that can be activated without direct contact is required.

In other words, the remote input using light or electromagnetic wave is suitable.

In the recent years, the use of biomaterials in electronic devices has been receiving a great deal of attention [7]. These materials offer diverse novel functions, are easy to fabricate into films, and are compatible with mechanically flexible substrates. One example is the photosensitive protein bacterioRhodopsin (bR), which has attracted interest in a variety of optical systems because of its diverse novel functions on light-signal processing, high sensitivity and stability to light [8,9]. bR is a nano-scale photoreceptive protein that works as a light-driven-proton-pump on the skin of \textit{Halobacterium halobium}. The nano-scale light-pumps in wild and other variable types of bR are reported to offer diverse light-signal processing functions including differential processing and memory function. For example, the differential response of bR was applied to an artificial photoreceptor which extracted moving objects from the image without signal processing [10,11]. Achieving the same functions with existing optical devices requires complex electronic circuitry for signal processing, as nano-scale structures similar to bR are difficult to construct with chemical synthesis or nanofabrication techniques.

In addition, bR is compatible with applications that require large area and mechanical flexibility, as it can be easily produced through bacterium cultures and coated on a substrate using low temperature processes [10,11,12]. However, this potential for large-area and flexible optical devices with bR has not yet been demonstrated, as past work focused on bR immobilization on inorganic ion-sensitive electrodes such as indium tin oxide (ITO) or tin oxide (SnO\textsubscript{2}). These
oxide electrodes require high-temperature annealing. Therefore, ion-sensitive electrodes which can be deposited at low temperatures are required.

In this paper we demonstrate that it is possible to replace the oxide electrode with the conducting polymer poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), making the photosensor element compatible with flexible substrates. Moreover, we integrate this new input device with an electrochromic display, making a device that responds to light-pen input by changing the color of the appropriate pixels. We discuss the individual components, as well as the fabrication process and the device characteristics.

2. Experimental

The overall device concept is shown in Fig. 1a. It consists of a flexible display with pixels that change color when activated by a light pen. The cross section of the device is shown in Fig. 1b. An electrochromic display based on PEDOT:PSS is combined with an array of photosensors based on a blend of PEDOT:PSS and bR. The two components were attached to each other so that their pixels were in registry (Fig. 1b). In order to understand device operation, it is important to examine the individual components first.

2.1. The photosensor component

A schematic of a test pixel for the light-pen input component is shown in Fig. 2a. PEDOT:PSS was blended with bR in solution and a film was cast on a polyethylene terephthalate (PET) substrate (see below for fabrication details). A poly(methyl siloxane) (PDMS) layer was used to define a reservoir containing a 0.1 mol/l potassium chloride buffer solution between the organic film and a Au counter electrode. The electrolyte is needed in order to activate the proton-pump function of the bR [13].

The pixel shows the typical differential response to light input from a green LED [10,11]. Namely, the photocurrent response consists of positive and negative transient currents when the illumination is switched on and off, respectively. This response can be understood as a modulation of the conductivity (and the resulting work function) of PEDOT:PSS. bR is known to generate protons upon irradiation, and this increase can de-dope the PEDOT:PSS, as the equilibrium [14]:

$$\text{PEDOT}^+ + \text{M}^+ + \text{e}^- \leftrightarrow \text{PEDOT} + \text{M}^+ + \text{PSS}^-$$

$$\text{(1)}$$

shifts to the right. In the above reaction M$^+$ are protons produced from bR. The transient current which is induced in the circuit reflects the equilibration to a new state as the work function of the PEDOT:PSS electrode changes. As soon as the illumination is turned off, bR absorbs protons. The opposite transient reflects the doping of PEDOT:PSS as the equilibrium in reaction (1) shifts to
the left. It should be noted that films of pristine PEDOT:PSS did not show any response to excitation with green light under the same conditions. It should be noted that the transient response of the current in Fig. 2b including the peak value of the current (about 2 nA) is the same order as the one of a BR photosensor with ITO electrode reported in previous studies [10,11]. The transient type of photocurrent is a unique feature, as most photosensors generate current continuously under steady-state illumination. Devices that employ such conventional photosensors detect the presence of light by measuring whether or not current flows in the photosensor. In contrast, the BR-based photosensor detects the presence of light by measuring positive and negative peak currents as illustrated in the Fig. 2b. The positive transient current fades upon continuous illumination (e.g. the light pen is parked over a pixel for a while), but the device uses the negative current associated with removal of illumination as an indication that the light pen has moved.

The response of the photocurrent to light intensity is shown in Fig. 2c and it is shown to saturate above 3 mW/cm². Typical laser pointers provide light intensities of 1 mW/cm², therefore, the sensor can be readily interfaced with commercially available light sources. The saturation is probably due to the saturation of BR response.

2.2. The display component

A schematic of a test pixel for the electrochromic display is shown in Fig. 3a. A PEDOT:PSS film was cast on an ITO covered glass slide (see below for fabrication details). A PDMS layer was used to define a reservoir containing 0.1 mol/l calcium chloride aqueous electrolyte. A PET window allowed optical transmission to be measured through the pixel, and an Al wire immersed in the electrolyte was used as the counter electrode.

Conducting polymers such as PEDOT:PSS are known to exhibit electrochromism [15]. When a negative bias of 1 V was applied to the PEDOT:PSS film, the color of the film with the thickness of 200 nm was switched from light- to dark-blue, as a result of increased absorption of visible light (Fig. 3b). This is in agreement with previous studies that have shown the reduction of PEDOT:PSS to be accompanied with increased absorption in the visible part of the spectrum [16]. Fig. 3c shows the light absorption of a pixel at 630 nm as the applied bias was alternated between −1 and +1 V at 200 mHz. The use of positive bias (+1 V) facilitates the oxidation of PEDOT:PSS and leads to a faster response of the display pixels.

2.3. Fabrication and integration of the two components

Since both the photosensor and the display components involve organic materials, patterning becomes challenging. Organic materials are not amenable to traditional photolithographic patterning, as several of the processes used in photolithography can damage them. Therefore, we patterned the organic layers using a modified process which involves a sacrificial parylene layer [17]. We also used an SU-8 layer, which can be developed with alcohol, and therefore be patterned on top of the organic layers without damaging them. The dimensions of the pixels were 1 mm², consistent with the spatial resolution obtained by the unaided human hand. Conductor lines to the pixels were made with PEDOT:PSS and had a width of 300 μm.

For the fabrication of both the photosensor array and the display components, the process starts by attaching a PET film (substrate) on a glass support with a UV curing adhesive for facile handling. Initially, PEDOT:PSS was patterned in an additive way using a parylene lift off process. Parylene (Parylene-C) was deposited on the PET film in a Lab Coater PD2010, to a thickness of 1.5 μm. Parylene was negatively patterned with SU-8 2025 (Kayaku Micro Chem) and etched by oxygen plasma at an oxygen flow rate of 5 ml/s (Samco corporation etcher) (Fig. 4a-1 and b-1). To enhance the conductivity of PEDOT:PSS (H.C. Stark), ethylene glycol was added into the solution at a volume ratio of 4:1 (PEDOT:PSS to ethylene glycol). The PEDOT:PSS was coated on the substrate at a spinning speed of 500 rpm and dried at the temperature of 75 °C for 15 min. The parylene layer was then mechanically peeled off, patterned the PEDOT:PSS film (Fig. 4a-2 and b-2). The substrate was subsequently heated to a temperature of 110 °C for 30 min, to dry the PEDOT:PSS film. Finally, an SU-8 layer was patterned on the organic material...
to serve as an insulation layer for the conductor lines (Fig. 4a-3 and b-3). Up to these steps the two components were nearly identical, with the only difference being a thicker PEDOT:PSS film (three coats) for the display component, to enhance color.

The fabrication of the photosensor component continued, with a deposition of parylene. The parylene was negatively patterned and etched. Then, a bR and PEDOT:PSS blend was coated. Wild type of bR (Munich Innovative Biomaterials) was added in purified water at a concentration of 20 mg/ml and sonicated for an hour. bR and PEDOT:PSS (containing ethylene glycol, as above) were blended at a 2:1 ratio and the blend was coated on a substrate. The film was dried under vacuum for 5 h, and was patterned by peeling off the parylene film (Fig. 4a-4 and a-5).

For the assembly of the photosensor array component (Fig. 4c), the PET substrate with the patterned layers was soaked with a 0.1 mol/l potassium chloride buffer solution and sealed with a blank sheet of PET. A gold wire was inserted between the two layers to serve as a counter electrode. The display component was assembled in a similar fashion using a 0.1 mol/l calcium chloride electrolyte and an aluminum wire counter electrode. Finally, the two components were glued together with an adhesive.

3. Results and discussion

The experimental setup is shown in Fig. 5. Excitation of the photosensor elements was achieved with a green LED (Philips’s Luxeon LXHL-MMCA). Each pixel was connected to a zero shunt current amplifier which consisted of an operational amplifier (OPA111), and a negative feedback resistor (10 MΩ). To amplify the signal and reduce noise, the output of the current amplifier was linked to an active low pass filter whose gain and cut-off-frequency were 40 dB and 100 Hz, respectively. The output voltage from a pixel was measured by a 16-bit analog to digital converter board (CSI-360116, Interface Corporation) and the light intensity was determined by the relationship between the photocurrent and intensity (Fig. 2c). The pixels of the electrochromic display were driven at a voltage of 1.0 V using a voltage follower circuit comprising of an operational amplifier (OP07). A digital input and output board (CSI-293116) was linked to the circuitry for turning the pixels dark- or light-blue according to the input from the photosensor array. The input and

![Fig. 4. Fabrication process for the photosensor array and the electrochromic display components, and device assembly. The two components were sealed with a PET film before assembly (not shown).](image1)

![Fig. 5. The experimental setup of demonstration of device operation.](image2)
Fig. 6. Demonstration of device operation. Input shapes (a transverse line, a square, the letter “H”), images obtained by the photosensor array, and projected images on the flexible display are shown.

Fig. 7. The bending test of the device. (a) Demonstration of device operation when bent to the curvature radius of 30 mm, (b) photosensitivity under bending, and (c) light absorption under bending.
output signals were controlled by a computer through the AD and Digital I/O boards.

Fig. 6 shows the input excitation shapes, the resulting images obtained by the photosensor arrays, and photos of the images on the display. Illumination patterns that consisted of a transverse line, a square, and the letter “H” (shown as the open pixels in the top-most figure) were drawn and successfully detected and displayed as light-blue pixels. Given the intensity dependent response of the photosensor array (Fig. 2c), the device was capable of recognizing illumination levels.

Since an electrochromic display was used the images were displayed as changes in the optical absorption of the PEDOT:PSS film (rather than in emission, as is the case of light emitting diode-based displays). Therefore, the process of displaying the information did not affect the sensitivity of the photosensor array. Furthermore we examined the functionality of the device when the substrate was bent. Fig. 7a shows the sensor input and display output when they are pasted on the curved surface with the curvature radius of 30 mm. The input and output of a transverse line (light blue pixels from top right to bottom left) were not affected when the device was bent. Fig. 7b shows the peak current from a 1 cm² photosensor under illumination at the intensity of 3 mW/cm² as a function of curvature. At negative values of curvature, the sensor is bent compressively. The data shows that the response of the photosensor is rather unaffected by bending. In a similar way, the coloration efficiency of a display pixel was examined when bent by monitoring the absorption of light with a wavelength around 630 nm as shown in Fig. 7c. The absorption did not change and bending did not break the pixels. Therefore, the device presented here can function when bent.

4. Conclusions

We demonstrated a novel light-pen input device with an integrated display. The input device consisted of a photosensor array made using blends of bacterioRhodopsin with the conducting polymer PEDOT-PSS. The display was electrochromic and was based on PEDOT-PSS. Patterning of the organic materials using a parylene technique was demonstrated. The use of organic materials allowed the entire device to be deposited on a thin plastic substrate and, therefore, to be mechanically flexible. The device successfully recognized various input patterns, even when bent. The integration of bR with conductive polymer will lead to large-area flexible optical devices: a overview, Biosensors and Bioelectronics 10 (1–2) (1995) 105–127.

Biographies

Seiichi Takamatsu is a doctoral course student in the Department of Mechano-Informatics of The University of Tokyo. His research interests are organic semiconductors.

Dr. Maria Nikolou received her Ph.D. in physics from the University of Florida in 2005. Her graduate work was on in-situ spectroscopic studies of single-walled carbon nanotubes and conjugated polymers in electrochromic devices. Her current research focuses on the development of biological sensors based on organic thin film transistors.

Dr. Daniel A. Bernards received a Ph.D. in materials science & engineering from Cornell University in 2007. His research involves the development of chemical and biological sensors based on organic thin film transistors.

John DeFranco is a graduate student in materials science & engineering from Cornell University. His research involves the development of organic thin film transistors.

Dr. Maria Nikolou received her Ph.D. in mathematics and physical sciences from the University of Groningen, the Netherlands in 1995. He is currently an Associate Professor in the Department of Materials Science & Engineering at Cornell University. His group’s research is in the area of organic electronics.

Dr. Kiyoshi Matsumoto received B.E., M.E., and Ph.D. degrees in mechanical engineering from The University of Tokyo, Tokyo, Japan, in 1985, 1987, and 1999, respectively. In 1987, he joined the Central Research Laboratory of Hitachi Co., Ltd., where he researched and developed optical disk systems. In 1995, he joined the University of Tokyo and is now a Professor in the Department of Mechano-Informatics, School of Information Science and Technology. His research interest is microsystems.

Dr. Isao Shimoyama received B.E., M.E., and Ph.D. degrees in mechanical engineering from The University of Tokyo, Japan, in 1977, 1979, and 1982, respectively. He joined The University of Tokyo in 1982 and is presently a Professor in the Department of Mechano-Informatics, School of Information Science and Technology. He has been a MEMS conference committee member since 1996, and his current research interest is microsystems, including MEMS structures and functions based on insects.

References


