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Polymer Light Emitting Diodes Powered via Paper-mounted Electronics

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Abstract— We have interfaced an array of polymer light emitting diodes (OLEDs) fabricated onto a glass substrate to a sheet of paper via a pressure sensitive conducting adhesive. By screen-printing a series of capacitive touch pads and connecting tracks onto paper using a low-cost conductive graphite ink, we are able to drive individual pixels in the OLED array via CMOS-based electronics that are also attached to the paper. Three AA batteries are used to power the CMOS electronics, touch-pads and the OLED array, with pixels in the array operating at a brightness of up to 210 cd/m^2. The work highlights a practical interface between plastic- and paper-based electronics.

Index Terms—Light emitting diode, capacitive touch pad, paper, electronics

WHILST paper is still widely used as a traditional print medium, there is growing interest in its use as a substrate for printed-electronics. [1] Paper has significant potential advantages compared to the plastic substrates often explored for printed electronics, as it combines low cost and easy recyclability. A wide range of different devices have been explored using paper-based substrates, with technologies such as transistors[2], displays[3], solar cells[4], batteries[5], supercapacitors[6], actuators[7] and sensors[8] all printed onto paper. Despite such remarkable progress, paper is not a suitable substrate for all types of electronics, as its high surface roughness and porosity together with high levels of chemical impurities make it incompatible with thin-film devices such as organic light-emitting diodes.[1] There is however considerable interest in the integration of thin-film light-emitting devices with paper and cardboard substrates, as this offers significant commercial opportunities in the high-value packaging for products such as perfumes and cosmetics etc. Some progress has already been made; organic light emitting diodes (OLEDs) have been fabricated onto a SiO_2 coated cellulose substrate coated with an ITO anode that was deposited using radio frequency magnetron sputtering, with a brightness of up to 3516 cd m^{-2} achieved at an applied voltage of 12 V.[9] Nevertheless, the high porosity of paper makes it a poor choice of substrate for OLED devices, as organic semiconductor devices have significantly reduced stability when exposed to oxygen or moisture.

To explore the use of paper as a substrate for OLED applications, we have used a pragmatic approach, and have used a paper substrate that has been screen-printed with low cost conductive carbon-based tracks. We show that such electronic tracks can deliver current to individual pixels in a polymer LED array that has been fixed to the paper surface using a conducting adhesive tape. Critically, the polymer LED array is fabricated using conventional techniques onto a thin, glass-substrate and is encapsulated with a glass cover-slip. This approach allows an air-stable, paper-mounted display to be realised without the problems associated with fabricating OLEDs directly onto paper. Here, we use a simple monochrome display consisting of an electroluminescent polymer. A practical embodiment of this technology would certainly require the glass substrate of the OLED to be replaced by a flexible plastic substrate, with the monochrome polymer display replaced by a multicolor-display incorporating high-efficiency low-mass molecular dyes. Nevertheless, our work demonstrates that organic displays can be easily interfaced to paper-based electronics, and such electronics can deliver sufficient current to drive OLEDs at a brightness to make simple information-displays that can be addressed by touch-pads.

Many consumer electronic devices are driven by touch -screen panels; indeed this now forms a key component in mobile communication devices and informative devices such as mobile cellular phones, navigation systems, informative flat panel displays, and mobile informative pads. [10] Touch-pads typically operate by detecting changes in resistance or capacitance, measure surface acoustic waves, or otherwise are based on infrared/optical detection. [11] Of these competing technologies, capacitive touch pads typically combine durability with accuracy [12] and have previously been fabricated using graphene oxide[13] or silver nanowire[14] inks on flexible plastics and paper substrates. Here, we print touch-pads from a carbon-based ink, and detect changes in the charge stored on the touch-pads as they are lightly touched using CMOS electronics that are also fixed to the paper. The CMOS electronics are then used to switch power to individual

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devices within the OLED array that is interfaced to the same paper substrate. In our demonstration device, the CMOS electronics (like the OLED display) are fixed to the paper substrate using a conductive tape. We have chosen to incorporate conventional silicon-based electronics with paper, rather than directly print transistor devices onto the paper substrate. This approach has the advantage that the electronic circuitry used is compact, reliable and has very low cost (potentially a few cents). Our work demonstrates the integration of paper electronics, low-cost capacitive touch pads and OLED technology and presents a new opportunity for the development of a range of consumer products.

The OLED arrays were fabricated onto 5 cm x 5 cm glass substrates coated with pre-patterned ITO that was sub-divided into 6 strips, each of which had a width of 2.54 mm. The substrates were sequentially cleaned by sonication in 10% (wt%) sodium hydroxide solution, Hellmanex solution, IPA and deionised water. Substrates were then dried using a jet of compressed nitrogen and baked at 120 °C for 5 minutes before use. PEDOT:PSS (HC Stark Clevios P VP AI4083) was filtered through a 0.45 μm PVDF filter before being spin coating at 5000 rpm onto the substrate to form a 30nm thick film. The device substrates were then thermally annealed at 120 °C for another 5 minutes to remove all moisture before being transferred into a glove box. A 5 nm layer of the hole-injecting co-polymer poly[(9,9-dioctylfluorenyl-2,7-diyl) -co-(4,4′-(N-(4-sec-butylphenyl)diphenylamine)] (TFB) (see chemical structure in figure 1(a)) was then spin cast onto the PEDOT:PSS from a chlorobenzene solution. The use of a TFB interlayer is known to significantly improve the performance of OLED devices.[15] Such improvements include enhanced efficiency and lifetime, as well as reduced turn-on voltage; effects that have been attributed to improved hole injection and transport [16, 17], and electron and exciton blocking[15, 18]. It is known however that a thin TFB layer can be dissolved during a sequential solution casting process.[19] To prevent this, substrates were thermally annealed at 150 °C for 30 minutes. This temperature is commensurate with the TFB glass transition temperature (T_g, around 150 °C [20]) and has been found to render it insoluble in organic solvents. An active layer of the light emitting co-polymer poly[(9,9-di-n-octylfluorenyl-2,7-diyl)-alt-(benzo[2,1,3]thiadiazol-4,8-diyl)] (F8BT) (chemical structure shown in figure 1(a)) was then spin cast onto the TFB from a toluene solution. F8BT is a prototypical light-emitting polymer that emits green fluorescence and can be fabricated into OLEDs having reasonable efficiency.[21]

A cathode consisting of 5 nm Ca capped with 100 nm of Al was thermally evaporated onto the F8BT layer through a shadow mask to form the cathode. The low work function Ca layer was included to facilitate electron injection into the F8BT layer, while the high reflectivity Al backplane helped increase optical outcoupling[22]. The cathode consisted of 6 stripes of width 2.54 mm^2 oriented at right angles to the ITO contact. The overlap between anode and cathode strips created an array of 36 pixels, with each pixel having an active area of 6.45 mm^2. To make a robust contact to the cathode, it was overlapped with an ITO pad at the edge of the substrate that had been wiped clean of all organic layers. This permitted a robust contact to be made to both anode and cathode contacts. To protect devices from the atmosphere, they were encapsulated using a 100 μm thick glass cover-slip via a UV-curable epoxy glue, with the encapsulation covering all of the device cathode. A schematic of the OLED stack is shown in Figure 1(b).

Before we describe the integration of the OLED devices with the paper substrates, we first discuss the performance of the OLED devices. A typical I-V trace recorded from a single pixel is shown in Figure 1(c) with corresponding L-V shown in Figure 1(d). Here, current was supplied to the OLED using a source measure unit, with luminance recorded with a Konica Minolta LS-100 luminance meter. We find that the device had a turn-on voltage of 2.2 V and reached a current density of 17,000 cd/m^2 at 8V. Control experiments demonstrated the importance of the TFB layer in obtaining a high brightness; it can be seen in Figure 1(c) and (d) that without the TFB interlayer, the device had a higher turn-on voltage of 2.4 V and reached a maximum luminance of 1100 cd/m^2 at 8V. We determine a maximum current and power conversion efficiency for the devices with (without) the TFB interlayer being 2.3 (0.2) cd/A and 1.2 (0.13) lm/W, respectively.

Figure 1(c) indicates that the current density of the OLED device incorporating a thermally annealed TFB interlayer is slightly higher before turn-on, but then reduces as the device starts to emit electroluminescence. Similar effects have been reported in previous work. [23, 24] It has been shown that the surface morphology, optical absorption and ionization potential of TFB annealed above T_g are unchanged. [24] Furthermore the average state of molecular order in a TFB film is also not likely to be substantially changed by such an anneal process, as TFB is an amorphous polymer. [20] It is likely therefore that on annealing, an intermixed layer of PEDOT:PSS and TFB is formed at the surface of PEDOT:PSS layer [25], and this intermixed layer may affect hole injection. Nevertheless, our results are consistent with the TFB interlayer acting as an electron/exciton blocking layer, with recombination most likely occurring around the interface between the predominantly electron-conducting F8BT polymer and the hole-conducting TFB. This electron blocking layer can be successfully used to balance the hole- and electron- currents in the F8BT layer and thereby improve efficiency. Although the efficiency of hole-injection appears slightly reduced here, devices that incorporate the TFB interlayer reach luminance that is 17 times higher is achieved without such a layer being present. We note that this finding is in agreement with a previous report that demonstrated that the luminance of an F8BT OLED device was increased by a factor of ten times using a cross-linked TFB interlayer. [26]

To construct the capacitive touch pads and the electronic tracks to deliver power to the OLEDs, we have used a proprietary conductive ink formulation that is based on carbon black and graphite dispersed into a carrier solvent together with surfactants and binders. This ink was deposited onto a

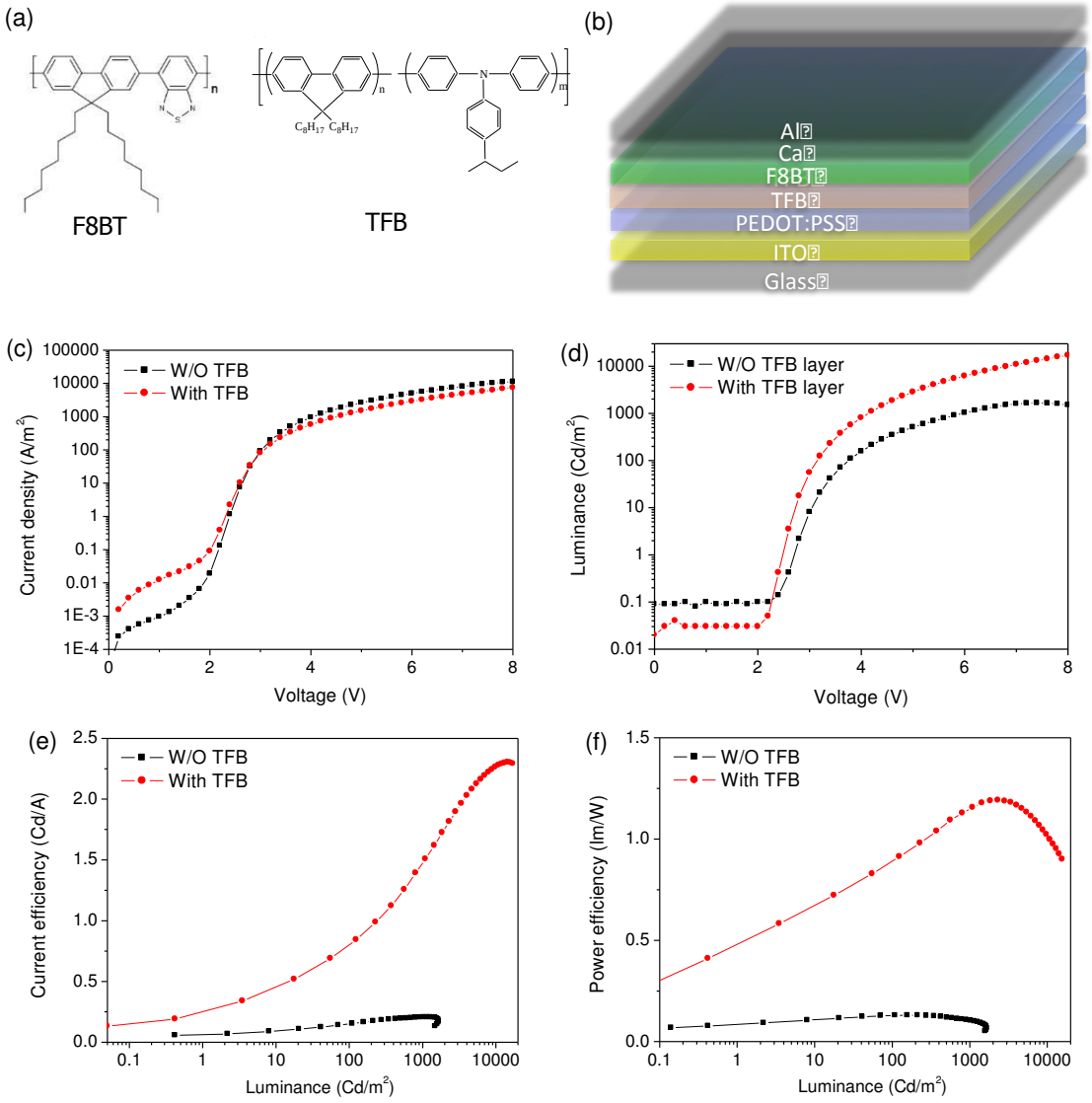

Fig. 1. (a) Molecular structure of the green light emitting polymer F8BT and interlayer polymer TFB. (b) Schematic cross-section through the OLED stack. (c) Current density and (d) luminance of the OLED device as a function of applied voltage with and without the presence of the TFB interlayer. (e) Current efficiency and (f) power efficiency of OLEDs as a function of luminance with and without the presence of the TFB interlayer.

220 µm thick paper substrate by screen printing forming a film having a thickness of around 7 µm. This ink was dried in air after printing and did not require additional sintering steps. Two SEM images of the printed tracks on the paper substrate recorded at different magnifications are shown in Figure 2. It can be seen that the printed material consists of an amorphous mixture of carbon black particles and graphite sheets, with surface scans using a surface profiler indicating that the layer has a root mean square roughness of around 2.35 µm over a distance of 3000 µm.

The layout of the capacitive touch pads and the tracks used to drive the OLED array are shown in Figure 3(a). Here, a 3 x 3 touch sensor array is used to control a 3 x 3 OLED array from the (6 x 6) OLED device via propriety CMOS electronics. The CMOS electronics board had a series of large contact pads on its underside that were interface to printed electronic pads on the paper surface using a conductive pressure sensitive adhesive tape supplied by 3M. The OLED array was similarly stuck to the surface of the paper using the conductive tape. The design of the track layout (see Figure 3(a)) permitted registry between the ITO tracks at the edge of OLED array and the conductive tracks to be made by hand. To mount the OLED display, it was flipped over so the thick glass substrate was uppermost. Electrical contact was then be made to the anode (1, 2 and 3) and cathode (A, B and C) strips via the few mm of ITO track that remained exposed at the edge of the glass substrate. Here, it was found necessary to place a small drop of silver-loaded ink (applied by hand) onto exposed region of the ITO before it was stuck to the paper using the conductive, adhesive tape.

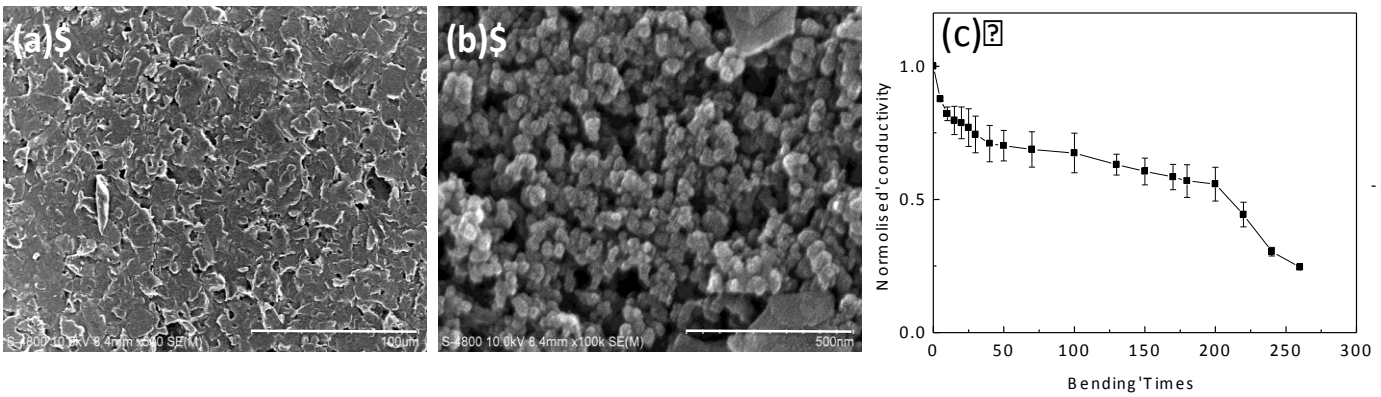

Fig. 2. (a) and (b) SEM images of the printed film at different magnifications. Scale bars are 100 μm and 500 nm for (a) and (b) respectively. (c) Normalized conductivity of the printed track as a function of blending cycle.

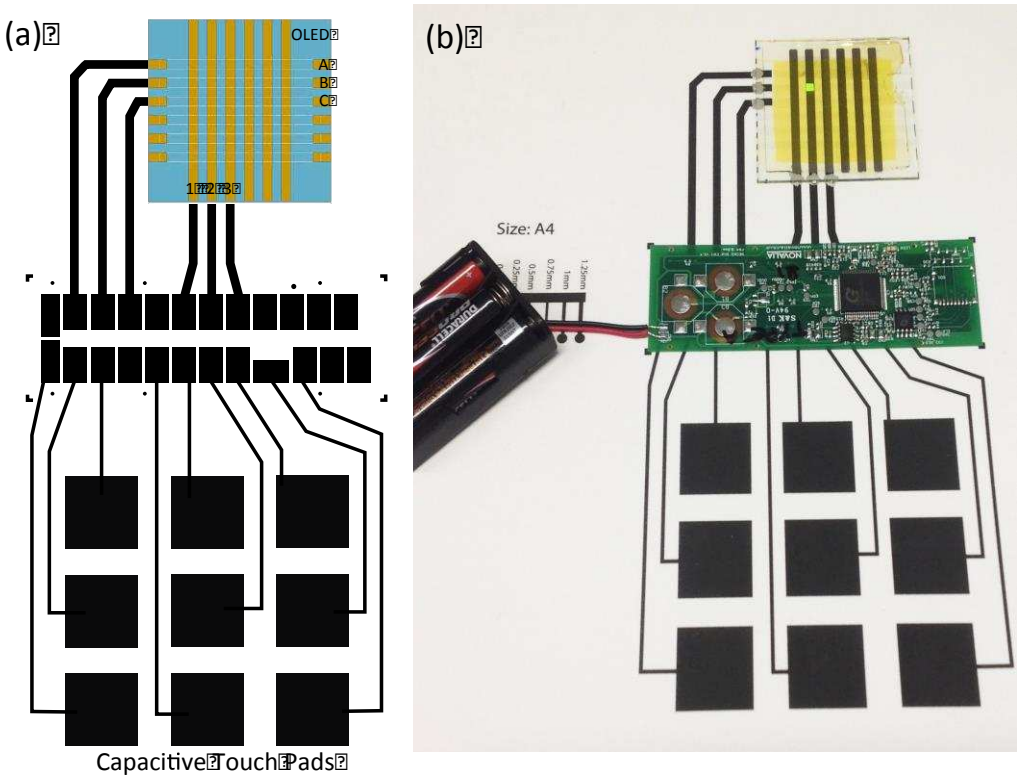

Fig. 3. (a) Schematic of the capacitive touch pad array, electrical circuit and LED array. (b) Photo of the integrated electronic system on the printed paper substrate. The three AA 1.5 volt batteries power the electronics and OLED array are clearly visible.

Briefly, the device works as follows. The CMOS electronics maintains a set-point charge on each of the capacitive touch pads. When a touch-pad is touched, the pad undergoes an instantaneous discharge that is then identified by the electronics. The on-board software then switched power to the appropriate pixel corresponding to the element on the touch-pad array that was activated. Power was provided to the switching electronics and to the OLED array by three AA 1.5 volt batteries, with device switching time being around 0.25 s. The "on" state can be switched off with another touch to the same or different pad. Gloved finger touch with nitrile gloves or with a finger works equally as well, with the switching of the OLEDs via the CMOS electronics following similar dynamics. Importantly, we found that there was not a notable voltage loss along the length of the printed tracks, as the voltage at the point of OLED contact was around 4.4 V. However, we found that there was a contact resistance between the printed track and the ITO track, with the voltage applied between the anode and cathode being reduced to approximately 3.4 V.

We have measured the luminance from individual pixels within the OLED array when powered using the system as shown in Figure 3(b) (with the photo presented taken under normal lighting conditions). We determine luminance values between 150 to 210 cd/m^2, with recorded luminance for the different pixels in the array summarized in Table 1. We find that the brightness of the brightest pixels is consistent with the luminance value shown in Fig. 1(d) for an applied voltage of 3.4 V. It can be seen that there is a systematic variation between

the brightness of the individual pixels within the array, an effect that appears to results from a voltage drop along the cathode strip. This effect could most likely be eliminated by using a thicker aluminium cathode strip to reduce series losses within the device.

TABLE 1
Luminance of individual pixels within the OLED array shown in Figure 3(b) when powered by three AA batteries via the printed electronic tracks.

Luminance (cd/m^2)	1	2	3
A	210	170	160
B	210	170	150
C	210	160	150

To evaluate the mechanical stability of the printed electronic tracks, we measured the relative conductivity along a 50 mm section of a 2 mm wide track after repeated bending cycles as shown in Figure 2c. Here, the paper was folded back on itself (-180°) around an 8 mm diameter cylinder, after which it was then bent in the opposite direction (again around a 8 mm cylinder) to +180°, with the bending process done by hand over 260 cycles. Here, the relative conductivity (normalized to the start of the experiment) was measured after each bend using a digital voltmeter to determine resistance, with the data presented averaged from 3 separate sets of bend-cycle measurements. It can be seen that over the first 25 bend cycles, the track looses almost 25% of its conductivity. This conductivity loss then slows, with the track conductivity reducing to 50% of its original value after 200 bending cycles. We believe that this mechanical stability is perfectly commensurate with the packing applications that we target (vide infra). We can instead create tracks having higher mechanical stability by printing thinner conductive inks between laminated polyester sheets. Such tracks can instead be subject to 100,000's of bend cycles without significant reduction in conductivity.

We have therefore created a simple demonstration of a plastic-electronic device being controlled via conventional electronics that are printed or attached to a paper–substrate. Here, the integration of technologies offers significant commercial opportunities. We note that paper based packaging industry is currently worth many $100bn, with further growth in this market anticipated. Here, the ability for a manufacturer to gain market share by being able to distinguish its products from that of its competitors is important, and will drive further innovation, design and development, which may well result in significant market pull into other areas. For example the combination of a cheap, ultra-low-cost and disposable flexible display and CMOS electronics that can also be produced for a few cents that could both be laminated onto cardboard or paper packaging could allow customers the ability to interact with their product in interesting and innovative ways. This could be used in simple greetings-cards or other products in which a customer could receive a simple message. This could be used as an effective way for a manufacturer to improve brand awareness, or otherwise include information (e.g. safety information for pharmaceutical products). More complex packaging applications could involve the use of displays that actively help the user obtain optimal performance from the product. For example a countdown-timer on a side of a packet displaying 'traffic-lights' could help a user control a time-dependent process and indicate when something was 'ready'. There are many products in which this could be useful, including paints, adhesives, hair-colorants and in home-baking. The most valuable application however we believe would be in products in which the value of the product is ultimately linked with to design and sophistication of the packaging. Examples of this include packaging for perfume, cosmetics and consumer-fashion.

In summary, we have integrated capacitive touch pads that have been screen-printing on paper substrates using low cost carbon-based inks with an OLED array via conventional CMOS electronics, with both the OLED array and the electronic board attached to the paper via a conducting adhesive tape. The on/off state devices in the array can be switched by simply touching a capacitive touch pad. The devices operate at a brightness of around 200 cd m^{-2}, with the system powered by 3 AA batteries (equivalent to 4.5 V). We discuss possible applications for this technology.

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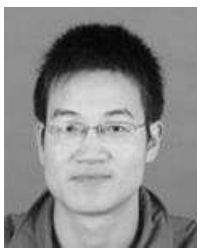

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