Organic Light Emitting Diodes and Photodetectors: Towards Applications in Lab-on-a-Chip Portable Devices

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ABSTRACT

We report that polymer light emitting diodes (pLEDs) and polymer photodetectors can be integrated on disposable polydimethylsiloxane [PDMS] microfluidic flowcells to form hybrid microchips for bioluminescence applications. PLEDs were successfully employed as excitation light sources for microchip based fluorescence detection of microalbuminuria (MAU), an increased urinary albumin excretion indicative of renal disease. To circumvent the use of optical filters, fluorescence was detected perpendicular to the biolabel flow direction using a CCD spectrophotometer. Prior to investigating the suitability of polymer photodiodes as integrated detectors for fluorescence detection, their sensitivity was tested with on-chip chemiluminescence. The polymer photodetector was integrated with a PDMS microfluidic flowcell to monitor peroxyoxalate based chemiluminescence (CL) reactions on the chip. This work demonstrates that our polymer photodetectors exhibit sensitivities comparable to inorganic photodiodes. Here we prove the concept that thin film solution-processed polymer light sources and photodetectors can be integrated with PDMS microfluidic channel structures to form a hybrid microchip enabling the development of disposable low-cost diagnostic devices for point-of-care analysis.

Keywords: polymer light emitting diode [pLED], photodetector, microfluidic chip, polydimethylsiloxane [PDMS], fluorescence, chemiluminescence, Lab-on-a-Chip.

1. Introduction

Since the concept of a Micro Total Analysis System (μTAS) or so-called Lab-on-a-Chip technology for integrated chemical and biochemical analysis was proposed in 1990[1], significant progress has been made in integrating various functional components into microfluidic analytical systems. The field has experienced ever expanding interest and research that has fuelled many different application areas including clinical point-of-care diagnostics [2], drug discovery [3], biological & chemical analysis [4][5] and forensic investigation [6]. This is due in part to the numerous advantages afforded by microfluidics such as low sample and reagent consumption, faster analysis, high throughput screening and automation. While polymer light emitting diodes (pLEDs) have been successfully developed for display applications in recent years, the use of pLEDs and photodetectors integrated with microfluidic chips for portable in-the-field or point-of-care (POC) devices represents a new field where the easy processing and low cost of the devices are advantageous.

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In the wider context of organic electronics research, pLEDs have been successfully employed in the display industry [7][8] while polymer photodetectors have been applied as solar cells and scanning sensors.[9][10] The use of pLEDs and polymer photodetectors as integrated light sources and detectors in miniaturised systems has recently attracted attention due to the wide range of accessible wavelengths and high luminosities of pLEDs, and good photo-responsivities of polymer photodetecors in the visible wavelength range. A group at Iowa State University in Ames and the University of Michigan in Ann Arbor have jointly reported a platform of fluorescent chemical and biological sensors, based on the structural integration of a fluorescent sensor with an organic light emitting diode [OLED] excitation source [11]. Another group at the University of Tokyo in Japan recently reported work using an OLED as an excitation light source integrated on a microfluidic chip to detect a fluorescence signal from dye labelled DNA and various proteins [12]. Here we report the use of solution processed thin film pLEDs as integrated excitation light sources on a microfluidic chip for albumin fluorescence detection and using a polymer photodetector to monitor on-chip chemiluminesence. The pLED and photodetector used for such analytical applications require sufficient stability and durability for quantitative testing. This has been achieved using a special packaging technique and chemically stable materials. PLEDs and polymer detectors are typically sensitive to moisture and oxygen and unwanted reactions between different layers of the device can occur [13]. To the best of our knowledge, we report for the first time the use of a solution-processed polymer thin film photodetector integrated with a microfluidic chip applied to bio analysis. Solution-processed thin film organic devices are compatible with roll-to-roll manufacturing on flexible plastic devices, thereby offering the potential of low-cost and high volume manufacture.

In our fluorescence detection work, we used a pLED as a novel light source to detect albumin on-chip. In this fluorescence assay, the analyte albumin was reacted with Albumin Blue 580 (AB580), a dye that binds selectivley to albumin with a concomitant increase in fluorescence. The emitted fluorescence signal was collected with a CCD spectrophotometer. The pLED made from a poly-phenylene vinylene derivative had strong yellow emission to match the absorption of AB 580. Our preliminary results show that signals can be detected at diagnostically relevant levels from 10μg/ml to 100μg/ml using the pLED as the excitation source. This sensitivity is sufficient for the determination of microalbuminuria (MAU), an increased urinary albumin excretion indicative of renal disease (clinical cut-off levels: 15-40 μg/ml).

In addition to the pLED light source fully integrated on-chip fluorescence detection also requires an integratable photodetector, we thus developed a polymer photodiode and characterised its performance by detecting chemiluminescence (CL) generated on-chip. Solution-processed photodetectors were made from blends of regio-regular poly(3-hexyl thiophene) [P3HT] and (6,6)-phenyl-C_{61}-butyric acid methyl ester [PCBM], which have a wide spectral response from 350nm to 700nm. For the CL testing we selected hydrogen peroxide as the model analyte since it is formed in many enzymatic diagnostic assays when in contact with analytes such as alcohol, glucose and cholesterol. The other active components used in the assay were bis(2-carbopentyloxy-3,5,6-trichlorophenyl) oxalate (CPPO), 9,10-diphenylanthracene dye and imidazole catalyst. Measurements of the CL spectra with analyte present showed a strong emission between 420 nm and 470 nm, which overlaps well with the photoresponse of our detector. A linear response between detected CL signal and hydrogen peroxide concentration was obtained from 10 μM to 1 mM. The results show a detection limit of ~10μM (S/N=3) which is similar to the reported detection limits for inorganic photodiodes [14]. This work demonstrates that our solution-processed semiconducting polymer photodiodes can provide a low-cost route towards sensitive, compact integrated micro analytical systems, such as needed for portable and disposable in-the field or point-of-care analysers for a variety of applications.

2. EXPERIMENTAL AND RESULTS

2.1 Design and Fabrication of polymer LEDs, photodetectors and microfluidic chips

A pLED is an electronic device made by sandwiching a series of organic or polymer thin films between two electrodes. Typically transparent indium-tin-oxide (ITO) is used as the anode and metal layers such as Ca/Al, LiF/Al, Ba/Al, Mg/Ag, are suited to use as the cathode. The dimensions of the individual pixels are determined by the spatial overlap of the cathode and anode. When electrical current is applied, a bright light is emitted through the transparent electrode i. e. the ITO side. This process is called electroluminescence. Even with the layered structure, these devices are very thin,

usually less than 500 nm. Patterning of the ITO is performed by standard photolithography. The conductivity of the ITO used here is 10 ohm/sq. In order to optimise effective excitation and signal detection, the geometry of the polymer LED and photodetector was designed so as to provide varied pixel sizes of 0.5mm x 0.5mm, 1mm x 1mm, 2mm x 2mm and 3mm x 3mm. The layout of the complete device including ITO anode and cathode is shown in Fig. 1. After ITO patterning, the surface is treated with an O_2 plasma to enhance the surface energy and make it more hydrophilic. A poly (3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) thin film was then spin-coated from aqueous solution to form a hole injection layer of 60-80 nm thickness on top of the ITO anode coated glass substrate. The resulting film was annealed at 140°C for 30 minutes in air to prevent water being trapped inside the film which can lead to degradation of the devices. The phenyl-substituted phenylene vinylene light-emitting copolymer solution was spin-coated on the top of the PEDOT:PSS layer to yield a ~40 nm film. Then the coated substrate was transferred into a glove box (dry nitrogen atmosphere) and heated on a hot plate at 110°C for 30 minutes. Thermal evaporation was then used for cathode deposition with a metal mask defining the pixel dimensions. 20nm calcium and 150nm aluminum layers were deposited on the polymer coated substrate in sequence. After evaporation, a thin UV curable epoxy layer was used to seal a metal can over the coated substrate. A desiccant patch was placed inside this package before sealing. The epoxy was cured by UV light. The completed device can be stored for years with negligible degradation due to the active layers being protected from moisture and oxygen [13].

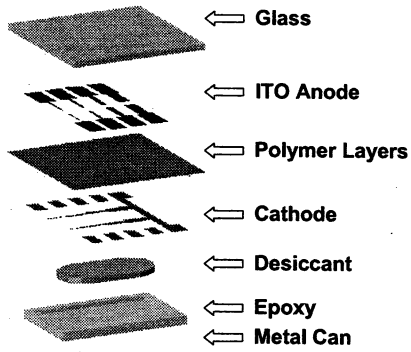

Fig 1 (a) The layout of the polymer LED and photodetector device structure

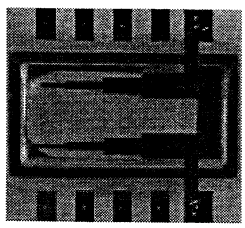

Fig 1 (b) The fabricated pLED device.

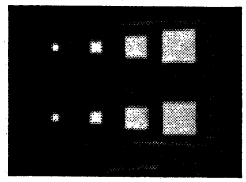

Fig 1 (c) Light emission from the pLED device.

Organic photodetectors were made using bilayer organic thin films sandwiched between an aluminium cathode and an indium tin oxide (ITO) anode. Poly(3,4ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT:PSS) was spin-coated from aqueous solution to form a hole injection layer of 60-80 nm thickness on top of an ITO anode coated glass substrate. After annealing at 140°C for 30 minutes in air, the P3HT:PCBM blend with 1:1 weight ratio in dichlorobenzene solution was next spin-coated on top of the annealed PEDOT:PSS film to a thickness of ~ 150nm. Aluminum was then thermally deposited through a shadow mask on top of the polymer layer to form a cathode of 200nm thickness. The completed devices were then further annealed at 130°C for 30 minutes in a nitrogen atmosphere to optimise the blend film morphology, thereby enhancing exciton dissociation and charge carrier transport characteristics. Devices with a range of areas, namely 0.25, 1, 4, and 9 mm^2 were fabricated and encapsulated in a nitrogen atmosphere using a UV-cured adhesive and metal or glass can structures fitted with desiccant patches. The structure of the photodetector devices is similar to the layout of the pLEDs shown in Fig. 1 (a).

The poly(dimethylsiloxane) (PDMS) microfluidic flowcells used in this work were fabricated by moulding from an SU-8 master on a silicon substrate. Fabrication of the two level SU-8 master was performed at the Centre for Integrated Photonics (Ipswich, UK) using standard SU-8 processing protocols. A Sylgard 184 Silicon Elastomer kit (Dow Corning, Coventry, UK) was used for PDMS moulding. Monomer and harderner were mixed at a ratio of 10:1 w/w, degassed for 30 min and then poured over the SU-8 mater. Three microscope slides were employed to define the side surfaces and the top of the PDMS microfluidic structure layer during curing. After curing at 95°C for 1 hour, the microscope slides were

removed and the ~ 3mm thick PDMS layer was peeled off the master. Access holes were punched through the PDMS layer at the microchannel ends and fused silica capillaries were inserted to serve as fluid reservoirs and interconnects. The layout of the chip is sketched in Fig 2. The device comprises two inlets, and a meandering mixing channel, a detection area and an outlet. To complete the device, the structured side of the PDMS layer is reversibly attached to the glass side of the PCBM:P3HT photodiode, with care being taken to align the detection area with the photodiode pixel.

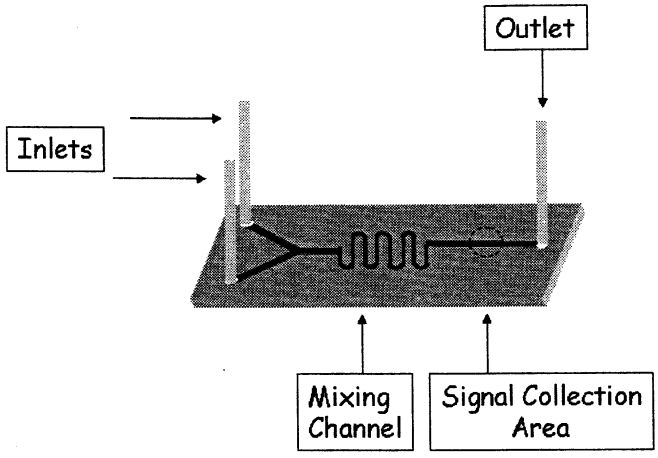

Fig. 2 The microfluidic chip layout used in this work.

2.2 Characterisation of pLEDs and photodetectors

The polymer LED was characterised using a 2400 Keithley source-measure-unit and Minolta 110 luminance meter. The current density against voltage and luminance against voltage characteristics are shown in Fig. 3. This yields important pLED operation parameters for the experiment. Here the pLED was current driven and brightness was monitored to compensate for possible device degradation, which is important for quantitative experiments and reproducible studies. The device efficiency curves are shown in Fig. 4.

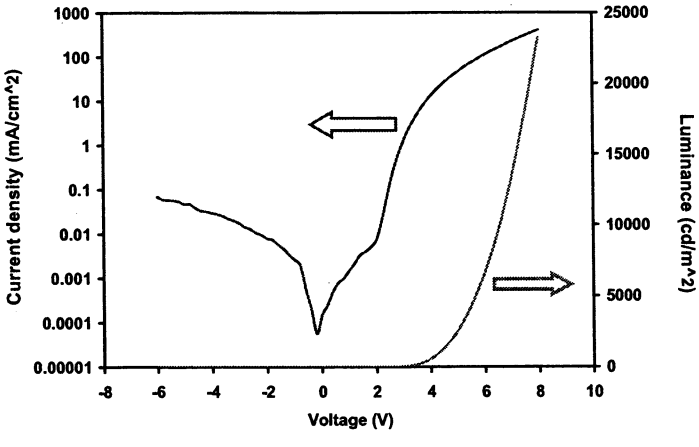

Fig. 3 The current density and luminance versus voltage characteristics of the pLED used in our studies.

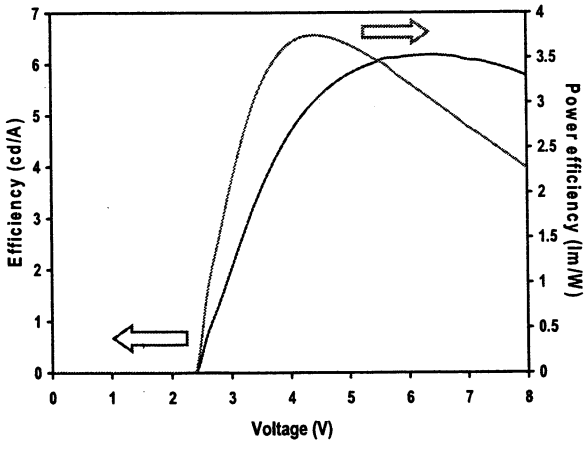

Fig. 4 The voltage dependence of the efficiency for the pLED used in our studies.

To characterise the photodetector, external quantum efficiencies (EQE) were determined as a function of wavelength with a 150 Watt Xenon Lamp (Bentham Instruments Ltd, Reading, UK) and a computer controlled CM110 monochromator (CVI Technical Optics, Onchan, UK). A Keithley 236 Source-Measure-Unit was used for the photocurrent measurement with a calibrated UV-enhanced silicon photodiode as reference (UV-818, Newport, UK). The EQE spectrum is shown in Fig. 5. The dark current density against voltage curve is shown in Fig. 6, measured by a Keithley 236 Source-Measure-Unit. The dark current of the detector used in this work is ~85 pA. The storage and operational lifetime of the photodetector devices are very good with no apparent degradation after more than one year storage and/or repeated use.

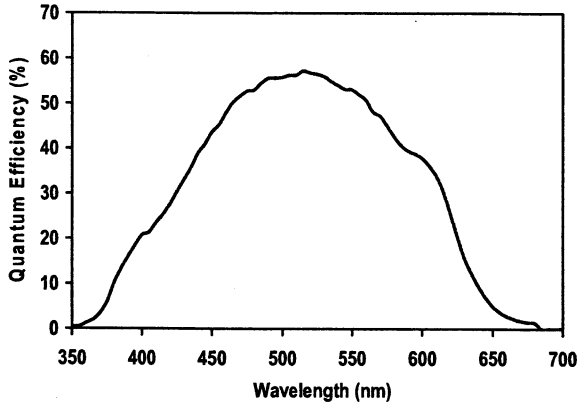

Fig. 5 The wavelength dependence of the external quantum efficiency of the photodetector used in our studies.

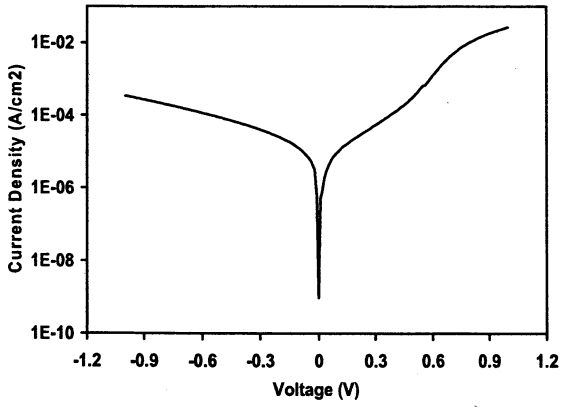

Fig. 6 The dark current density against voltage for the photodetector used in our studies.

2.3 Results for fluorescence detection of microalbuminuria (MAU)

All solutions were prepared from the Albumin Fluorescence Assay kit 09753 (Fluka Chemicals, Gillingham, UK) following the protocol provided in reference [15]. In short, AB580 stock solution was diluted 1:50 v/v in buffer, resulting in ~1.2 μM AB580. Human serum albumin (HSA) was dissolved in calibrator solution. AB580 and HSA solutions were hydrodynamically pumped through microchip inlets 1 and 2, respectively, at a ration of 2:1 v/v. The flow rate was 20μL/min. In initial experiments the detection set-up was optimised for maximum fluorescence signal from AB580/HSA. The overlap between the pLED emission and the absorbance band of the AB580/HSA complex is depicted in Fig.7. In the microchip experiments, the excitation light from the pLED was focused onto the detection area in the microfluidic chip using a focusing lens. An aperture was used before the chip to limit the scattered light from the light source. To circumvent the use of a longpass emission filter, an orthogonal detection geometry was successfully implemented. A CCD spectrophotometer equipped with an optical fibre was used to collect the HSA complex emission through the side surface of the PDMS microchip. The pLED was operated at 20 mA (constant current mode), yielding a brightness of ~8,000 cd/m^2.

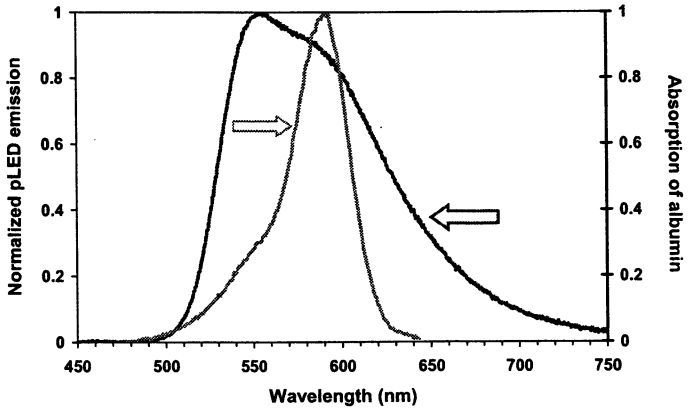

Fig. 7 The emission and absorption spectra of pLED and HSA solution, respectively.

For first quantitation experiments HSA and AB580 were mixed off-chip and then introduced onto the microchip for detection. HSA concentrations 1-1000 μg/ml were investigated, with water and AB580 dye serving as references. Fig. 8 depicts the smoothed and normalised data as recorded with the CCD spectrophotometer. A general increase of the HSA/AB580 complex emission peak at 620 nm can be observed for HSA concentrations of 30 μg/ml and higher. The normalised spectra reveal a discernible change of the complex emission peak for HSA concentrations as low as 10μg/ml. Also shown are the spectra recorded in the presence of water (pLED emission alone) and AB580 in aqueous solution (1:50) in the absence of the analyte.

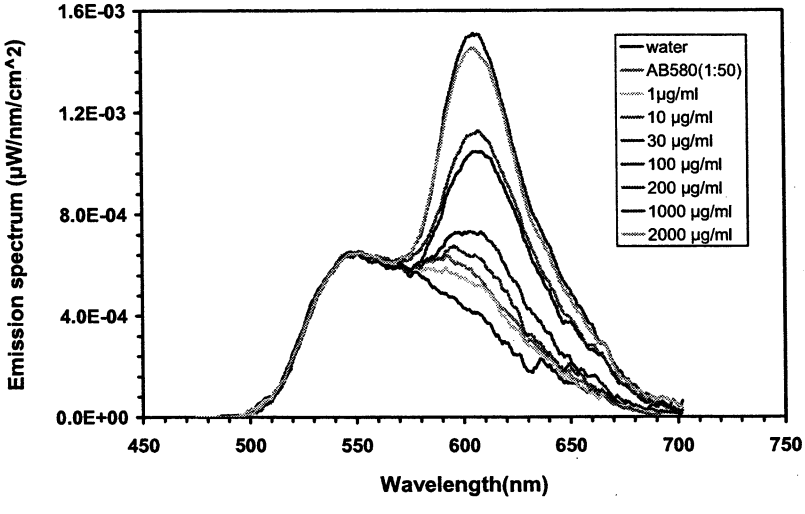

Fig. 8 The fluorescence emission spectra for different concentrations of HSA together with the spectra recorded for water and AB580 in water (no analyte).

Quantitation of the fluorescence intensity was achieved through integration of the spectra from 500 to 700 nm shown in Fig. 9. A strong signal increase can be observed for lower HSA concentrations followed by leveling-off and saturation. This is due to the HSA/AB580 complex formation obeying the mass action law and is in agreement with previously published calibration data for HAS [16]. The employed high AB580 excess is responsible for the steep calibration curve increase at low HSA concentration. Linear regression analysis shows a linear range from 1-100 μg/ml. HSA. In

subsequent experiments the effect of on-chip mixing on quantitation results was investigated. Initial experiments were conducted with flow rates of 25 and 50 µL/min for HSA and AB580, respectively. While the detected signal intensities were generally in the same range as for the complex mixed off-chip, a signal increase was observed for the first ~3 min after stopping the flow. This points to incomplete mixing due to an insufficient on-chip residence time of the reagents. Consequently the applied reagent flow rates were lowered to 10 and 20 µL/min, respectively. This completely negated the stopped flow effect, indicating now complete intermixing of the two reagents. At the optimised flow rates an average on-chip residence time in the mixing channel / detection chamber of ~122 s can be calculated. For AB580 this corresponds to an effective diffusion distance of ~110 µm (based on a diffusion coefficient of $5x10^{-11}$ m^2s^{-1}). Interestingly this is only less than one-third of the half channel width.

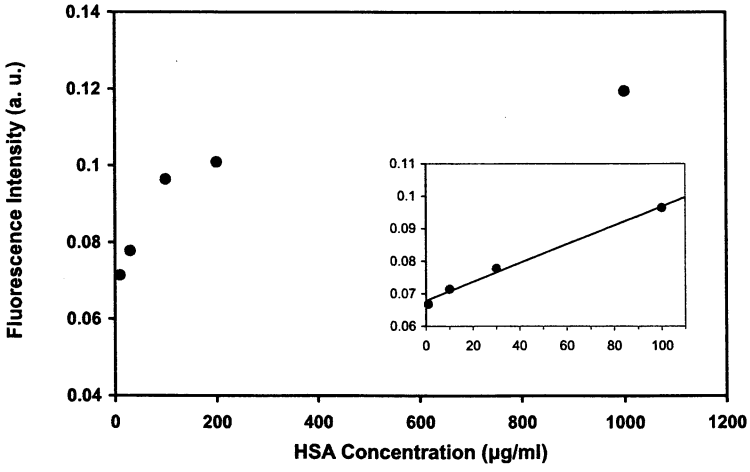

Fig. 9 The fluorescence intensity of the HSA/AB 580 complex as a function of the concentration of HSA. The inset shows the lower HAS concentration regime on an expanded scale together with a linear fit (R^2=0.997).

Adsorption of HSA inside the hydrophobic PDMS microchannels was subsequently investigated. since HSA loss could result in *false negatives* in MAU determination. Carry-over between runs was tested by first running the assay with high HSA concentrations followed by a run with AB580 only. Any HSA remaining in the microchannel should be complexed by the dye and thus be detectable as an increase in the complex emission peak at 620 nm. In between the runs the microchannels were flushed with water for 10 min at 20 µL/min. We generally observed that the recorded signals recovered to baseline level, indicating that there was *no* residual HSA in the microchannels after rinsing. While inter-run carry-over can be eliminated, some HSA adsorption during the assay was still likely to occur due to the high affinity of hydrophobic proteins to PDMS. A possible reason for the limited adsorption observed is oxidation of the PDMS channels during earlier chemiluminescence experiments. Oxidation of the PDMS results in a more hydophilic surface less prone to protein adsorption.

While the presented results clearly demonstrate successful MAU determination on a microchip, future efforts will focus on a more efficient coupling of the pLED light source to the microchip. This could be achieved through size matching of pLED pixel size and detection area, microcavity pLEDs with more confined light output, or the use of integrated PDMS microlenses.

2.4 Chemiluminescence detection

An additional study was used to investigate the potential of polymer photodetectors integrated on the PDMS microfluidic device to detect chemiluminescence (CL). Polymer photodetectors made from blends of regio-regular poly(3-hexyl thiophene) [P3HT] and (6,6)-phenyl-C_{61}-butyric acid methyl ester [PCBM] have a photoresponse that extends from 350 nm to 670 nm, with an external quantum efficiency $\geq$ 55% at 500 nm. Hydrogen peroxide formed in enzymatic diagnostic assays is selected here as a test analyte for CL quantification within the microfluidic device. The other active components used in the assay are bis(2-carbopentyloxy-3,5,6-trichlorophenyl) oxalate (CPPO), 9,10-diphenylanthracene

dye and imidazole catalyst. Measurement of the CL spectra with analyte present showed strong emission between 420 nm and 470 nm, which overlaps well with the photoresponse of our detector. An extensive study was performed to optimise the photodetector configuration for the best detection sensitivity of the peroxyoxalate based CL reaction.

A typical photodetector response for 500 μM hydrogen peroxide reacted on-chip with the CL reagents is shown in Fig. 10. Recording of the photodiode signal is started before initiation of the CL reaction, yielding a low level background signal. The reaction is started by simultaneously pumping hydrogen peroxide, CL reagent, dye and imidazole catalyst through the inlets of the microfluidic chip. Under the laminar flow conditions encountered in the mixing channel, the reagents mix by diffusion only. The degree of mixing can conveniently be controlled via the applied flow rate which defines the available mixing time on chip. Complete mixing results in the generation of a strong CL emission, which can be seen as an increase in the photodiode response. After the flow is stopped, the photocurrent from the detector drops back to the background noise level. For quantitation, different concentrations of hydrogen peroxide were injected into the CL reagent stream. A linear relationship between the photosignal and hydrogen peroxide concentration was obtained over a range of concentrations from 10 μM to 1mM (Fig. 11). Preliminary results show that the detection limit (S/N=3) for the detector is below 10 μM, representing over 100 times improvement compared to our previously reported results for a vacuum deposited small molecule CuPc/C_{60} photodiode [17]

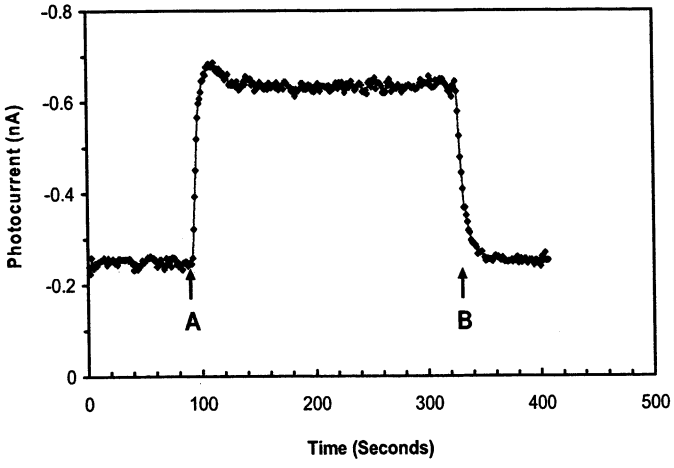

Fig. 10 A typical photocurrent response arising from the CL reaction on- chip. The regents were introduced at time A and the flow was then stopped at time B.

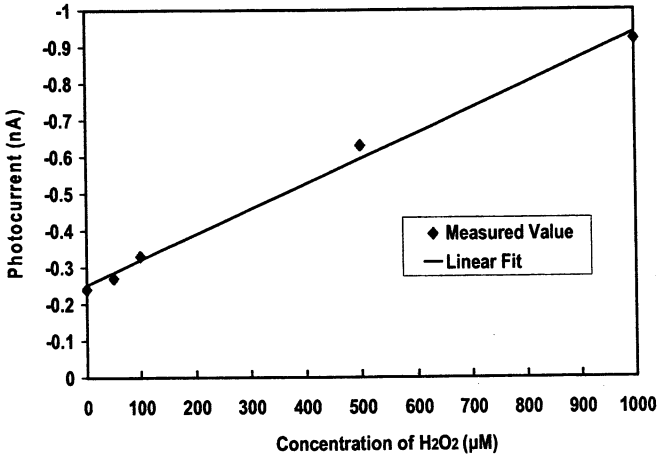

Fig. 11 The linearity of the photocurrent as a function of hydrogen peroxide concentration. The straight line is a linear fit (R^2=0.997).

DISCUSSION AND CONCLUSION

This report has demonstrated that polymer light emitting diodes can be employed as integrated light sources for fluorescence detection in portable Lab-on-a-Chip microdevices. As a first step towards a disposable diagnostic microchip for HSA determination we have reported the use of a thin-film pLED as an integrated excitation source for microscale fluorescence detection. The thin-film pLED can be easily fabricated on rigid or flexible planar substrates, facilitating integration with microdluidic devices. In a fluorescence assay, HSA was reacted with AB580, generating a strong emission at 620 nm when excited by a pLED with peak emission at 550nm. Filter-less discrimination between excitation light and generated fluorescence was achieved through an orthogonal detection geometry. When the assay is performed in a PDMS microchip at flow rates of 20 μL/min, HSA concentrations down to 10 μg/ml could be detected with a linear range from 10 to 100 μg/ml. This sensitivity was sufficient for the determination of microalbuminuria (MAU), an increased urinary albumin excretion indicative of renal disease (clinical cut-off levels: 15-40 μg/ml). As a further step towards a fully integrated detecion device with both the light source and photodetecor on-chip we fabricated and tested

an organic photodetector. The sensitivity and spectral response of the photodiodes were tested by detecting the emission of a chemiluminescence reaction performed on chip. We could demonstrate that our organic photodetectors exhibit sensitivities comparable to inorganic photodiodes. For integrated fluorescence detection on chip, a collinear geometry is often employed with the light source and photodetector on either side of the microfluidic device. For such geometries, short pass and long pass optical filters would be helpful. While the short pass filters can sharpen the pLED emission, the long pass filters on the detector side help to block excitation light from saturating the detector. Both short pass and long pass filters are currently being developed in our lab, bringing us one step closer to a fully integrated disposable on-chip fluorescence detection system, which could be invaluable for point-of-care diagnostic applications

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