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Fibre coupled micro-light emitting diode array light source with integrated band-pass filter for fluorescence detection in miniaturised analytical systems

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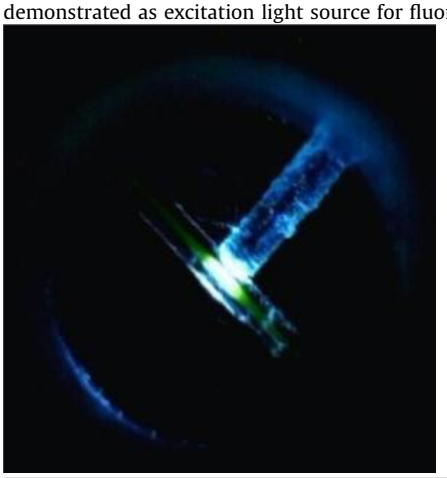
HIGHLIGHTS

- A new integrated miniaturized fibre-coupled solid-state light source is presented.
- Based on a micropackaged micro-fabricated light emitting diode micro-array (μ-LED).
- Interference filter micropackaged with optical fibre and the μ-LED array.
- Demonstrated as excitation light source for fluorescence detection.

GRAPHICAL ABSTRACT

A new generation of integrated miniaturized fibre-coupled solid-state light sources based on microfabricated light emitting diode micro-array (μ-LED), micropackaged with a custom band-pass optical interference filter deposited at the end of an optical fibre, is presented in this work and demonstrated as excitation light source for fluorescence detection in capillary electrophoresis.



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ABSTRACT

In this work, a new type of miniaturized fibre-coupled solid-state light source is demonstrated as an excitation source for fluorescence detection in capillary electrophoresis. It is based on a parabolically shaped micro-light emitting diode (μ-LED) array with a custom band-pass optical interference filter (IF) deposited at the back of the LED substrate. The GaN μ-LED array consisted of 270 individual μ-LED elements with a peak emission at 470 nm, each about 14 μm in diameter and operated as a single unit. Light was extracted through the transparent substrate material, and coupled to an optical fibre (OF, 400 μm in diameter, numerical aperture NA = 0.37), to form an integrated μ-LED-IF-OF light source component. This packaged μ-LED-IF-OF light source emitted approximately 225 μW of optical power at a

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bias current of 20 mA. The bandpass IF filter was designed to reduce undesirable LED light emissions in the wavelength range above 490 nm. Devices with and without IF were compared in terms of the optical power output, spectral characteristics as well as LOD values. While the IF consisted of only 7.5 pairs (15 layers) of SiO_2/HfO_2 layers, it resulted in an improvement of the baseline noise as well as the detection limit measured using fluorescein as test analyte, both by approximately one order of magnitude, with a LOD of 1×10^{-8} mol L^{-1} obtained under optimised conditions. The μ-LED-IF-OF light source was then demonstrated for use in capillary electrophoresis with fluorimetric detection. The limits of detection obtained by this device were compared to those obtained with a commercial fibre coupled LED device.

1. Introduction

Photometric and fluorimetric optical detection methods are frequently used in capillary based separation techniques including capillary electrophoresis (CE) and capillary liquid chromatography (cap-LC) [1–6]. While photometric detection is generally valuable and the most common detection in CE and cap-LC, the combination of the most sensitive detection method of laser-induced fluorescence (LIF) detection with CE provides a powerful separation platform with a wide range of advantages including speed, high resolution, efficiency, and sensitivity, as well as low sample and reagent consumption [7–9], for applications such as glycomics [10–12].

Traditionally used excitation sources for fluorimetric detection are incandescent or arc lamps (halogen or mercury) based on technologies going back over a century, and in the last decades on lasers and then increasingly on solid state light sources – diode lasers and LEDs [13–18]. Arc and incandescent lamps have an advantage in their broadband continuous emission; however, due to their size, fragility, heat production, relatively low luminosity and optical output stability, they are not suitable for miniaturization purposes. Lasers are commonly used as excitation sources due to their high emission intensity, monochromaticity and advantageous spatial properties (collimated light, easy to focus), which allow the light to be focused to a very small area. Light-emitting diodes (LEDs) since their discovery in 1907 [19] and commercial technology developments from 1960s pushed down the wavelength scale from infrared and red to green, blue, violet and ultraviolet [20–23], and are nowadays considered as the light sources of the future. LEDs offer numerous advantages including quasi-continuous wavelength coverage, stable intensity, robustness including long lifetime, small size, low cost, and ability to be pulsed at fast rates, while their main deficiency is the lack of powerful enough emitters in the deep-UV (below 300 nm) spectral region [24–26].

In the area of on-capillary detection including CE, LEDs have been used in miniaturised low-cost detection systems, both photometric [27–38] and LED induced fluorimetric (LED-IF) [39–43], with advantages especially for portable CE instruments [35]. A number of LED-IF detection designs for microfluidic chip-based CE [44–47] systems have been reported as well.

As LEDs, which otherwise would be more popular as miniaturised light sources for portable devices, are semi-monochromatic and naturally possess bandwidth of approx. 20–50 nm, when used as excitation sources in LED-IF for optimal performance and low baseline noise they have to be combined with low-pass filters [43,48,49].

Miniaturization of the individual optical components (light source, optical filters, lenses, mirrors, etc.) and their assembly into a functioning optical system is the limiting factor when creating a miniaturized CE-LIF design either LIF or LED-IF. As LEDs have wide spatial light distribution, focusing optics is usually required for optimal sensitivity of LED-IF detection [43,50,51]. Optical fibres directly coupled to LEDs (pigtailed LEDs) [44,52,53] are a very popular alternative in creating a spatially directed LED light source. Fibre-coupled LED sources with an integrated interference filter could be an attractive integrated micropackaged fibre-coupled light source component for miniaturised optical detection systems.

The μ-LED arrays [54] provide a quasi-collimated light emission and therefore can have a good coupling efficiency to optical fibres. When integrated and micropackaged with an optical fibre, an interference filter can be inserted between the μ-LED array and the fibre, by depositing this filter on the back surface of the substrate emitting LED. Such LED-based integrated and micropackaged optical fibre light sources emitting from the fibre spectrally filtered light, could become a new option in custom designed optical fibre-coupled light sources for fluorescence detection in on-capillary and microfluidic chip separation systems. The authors to their best knowledge are not aware of any other similar work on integrated micropackaged fibre-coupled μ-LED array light sources.

In this work, for the first time an integrated and micropackaged μ-LED array with deposited SiO_2/HfO_2 interference filter and coupled to an optical fibre (μ-LED-IF-OF) was designed, fabricated, characterised and demonstrated using CE as an excitation light source for capillary separation techniques with fluorimetric detection.

2. Experimental

2.1. Materials

For the microfabrication of the LED micro-arrays, GaN substrate material was purchased from LUMILOG (Sophia Antipolis, France). On the top of this substrate, epitaxial InGaN layers were deposited using MOVPE (metal organic vapour phase epitaxy) at the University of Cambridge (UK). Device processing and deposition of the integrated filter onto the back side of the LED wafer material were carried out in the cleanroom facilities of the Tyndall National Institute in Cork (Ireland). The optical fibre was purchased from Thorlabs (Ely, UK). The fibre had a core diameter of 400 μm and a numerical aperture of 0.37 (part no. BFH37-400). A perforated silicon platform was used to integrate the μLED chip with the optical fibre. This component was also fabricated in the cleanroom facilities of the Tyndall National Institute in Cork (Ireland).

The optical transmission spectrum of the glass slides was measured using a white light source and an Ocean Optics USB 2000 spectrometer (Ocean Optics, Dunedin, FL, USA).

2.2. Chemicals

Hafnium oxide (HfO_2) was purchased from PI-KEM (Tamworth, UK). Fluorescein and sodium phosphate were purchased from Sigma–Aldrich (Dublin, Ireland), ammonium acetate, acetic acid, and 8-aminopyrene-1,3,6-trisulfonic acid (APTS) were obtained from Sigma–Aldrich (St. Louis, MO, USA). A solution of 1×10^{-7} mol L^{-1} fluorescein was prepared in a sodium phosphate buffer (20 mmol L^{-1}, pH 9).