ABSTRACT

We present a portable microsystem to quantitatively detect cocaine in human saliva. The chip combines a simple microfluidic method for multiphase liquid-liquid extraction to transfer cocaine from IR-light absorbing saliva to an IR-transparent solvent with the on-chip cocaine detection by IR-waveguide-spectroscopy (QC-laser, waveguide, detector). With our droplet-based extraction method we achieve an additional analyte pre-concentration of at least two orders of magnitude.

KEYWORDS:

cocaine detection, liquid-liquid extraction, IR-spectroscopy, droplet generation, droplet merging

INTRODUCTION

On-site drug testing of easily accessible body fluids has gained a lot of importance both for road safety and forensic applications. IR-waveguide-spectroscopy was chosen as detection method. Unfortunately the characteristic absorption peak of cocaine is at 5.8µm - a wavelength where saliva (water) is strongly absorbing light as well [1]. Therefore, the multiphase liquid-liquid extraction system presented here brings the analyte to a transparent medium before detection.

H-filters, the common method for microfluidic liquid-liquid extraction [2], were tested as a reference system for our droplet based approach. To increase the diffusion interface it has been suggested to replace the parallel flow (H-filter) by a droplet-based approach with channel-wide droplets [3]. With our new extraction system we go a step further and enhance the extraction efficiency by producing smaller droplets and varying the flow ratio of the two liquids to allow pre-concentration. Our work also includes IR-absorption measurements of cocaine integrated on micro-chip level. To the best of our knowledge, this is the first time that such a measurement has ever succeeded.

EXPERIMENTAL

Our chip consists of an optical detection waveguide made of germanium on silicon (Figure 1), which was plasma-bonded to the microfluidic part (Figure 2). The mid-IR waveguide was made from a mono-crystalline germanium layer with standard photolithography and reactive ion etching by CF4. The microchannels were made by molding the UV-curable adhesive NOA81 (Norland Optical Adhesives) on a Scotch-tape master (Figure 3&4a). NOA81 was found to be a suitable material for low-cost forensic applications due to its low cocaine adsorption and chemical compatibility with organic solvents [4,5]. Figure 2 shows the complete optofluidic system including the connectors for the tubing.

Figure 5 provides an overview of the extraction system. First monodispersed perchloroethylene (PCE) droplets were generated in saliva using the capillary focusing effect (Figure 6) [6]. The depth of the channel mainly defined the droplet size, and allowed us to independently adjust the flow rates in order to reach the requirements for coalescence further downstream of the chip. After the extraction section, a shallow and hydrophilic side channel drained out the saliva. Coalescence of the PCE droplets was based on both compressive merging (compression of the droplets against the end of the extraction channel) and decompressive merging when the droplets are accelerated (constant flow rate, smaller cross-section) into the shallower hydrophobic PCE outlet-channel. Acceleration resulted in local droplet separation and induced the formation of two facing nipples in the contact area (local low-pressure). This brought the interfaces close enough to merge [7]. Finally, the cocaine extracted to the IR-transparent PCE was then measured in continuous phase by the optical waveguide.
RESULTS AND DISCUSSION

Droplets were generated using the capillary focusing effect [6]. When a slug-flow of PCE surrounded by saliva flowed over a step, PCE droplets were ripped off the slug. The droplet size mainly depended on the channel depth, showing a linear dependence (Figure 7). The generated PCE droplets were monodispersed showing variations of the diameter of less than 3%.

The presented droplet-based extraction system achieves up to two orders of magnitude better extraction efficiencies than the state-of-the-art H-filters. Depending on the imbalance of the flow rates we even manage to pre-concentrate the cocaine in PCE (Figure 8).

Quantitative on-chip measurements are feasible with a limit of detection (LOD) of 100µg/mL. Figure 9 shows the IR-waveguide-detection of different cocaine concentrations flowed (50µL/min) through a test device consisting of a short optical waveguide covered by a simple straight microfluidic channel. For the design and the description of the system refer to [8]. Figure 10 presents the detection of 500µg/mL cocaine spiked in saliva and extracted on-chip to PCE by using the presented droplet-based liquid-liquid extraction system.

CONCLUSION

The presented droplet-based microfluidic system enables up to two orders of magnitude better extraction efficiencies compared to the state of the art H-filters. Cocaine extracted from human saliva to PCE was detected by IR-waveguide-spectroscopy on the integrated optofluidic device. Our LOD of 100µg/mL is not sufficient to detect the saliva concentration of a single cocaine dose, which can exceed 1900ng/mL cocaine in human saliva for 2-4 hours [9], but we...
are working on improving our LOD by using smaller droplets (higher surface-to-volume ratio), more imbalanced flow ratios, a longer and wider extraction section (increased extraction time), and temperature stabilization of the laser (noise and baseline-drift reduction).

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