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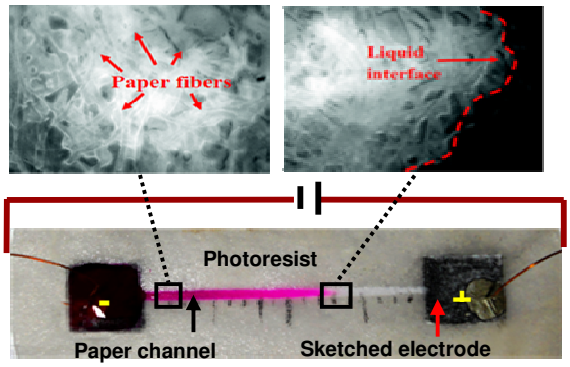

A ‘paper-and-pencil’ based inexpensive microdevice results in appreciable augmentation in the rate of microcapillary filling, by using electrokinetic phenomenon.

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Electrokinetics with 'paper-and-pencil' devices

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We demonstrate the occurrence of electrokinetic phenomenon in paper substrates, by developing a simple 'paper-and-pencil' device. The underlying electrokinetic phenomenon results in enhanced liquid transport through the paper-fibre matrix, which exhibits significant active electrical controllability and improved repeatability. These bear far-ranging consequences towards opening up a new paradigm of fluidics over small scales.

The recent advent of paper-based microfluidic devices, which are cost-effective, miniaturized, and circumvent the need for operational expertise, has greatly revolutionized the field of point-of-care diagnostics, especially in the developing countries[1]. The primary advantage of the paper-based devices stems from the fact that intricate two-dimensional and three-dimensional microscale patterns can be economically and efficiently fabricated on the paper substrate, for performing various analytical functions in tandem[2-4]. In these paper-based devices, the liquid transport is governed by the natural capillary action of the test liquid through the numerous microscopic flow passages formed by the paper-fibre arrangement[5], as pre-fixed by the paper manufacturing process. Hence, the natural transport of liquid through paper substrates is non-controllable, and has poor repeatability. To address these issues, successful attempts were made, very recently, to control, and manipulate, liquid flow in paper by using surface acoustic waves (SAW)[6], and by exploiting centrifugal force on rotating platforms[7]. To add a new dimension to the flow control techniques in paper-based devices, we demonstrate here, for the first time, the modality of electrokinetic flows in paper-based platforms. We propose a novel, yet elegantly simple, 'paper-and-pencil' microfluidic device for active electrical control of the liquid transport, by exploiting the occurrence of spontaneous electrokinetic phenomena in paper substrates.

Whatman® cellulose filter papers (Grade 1) are used for fabricating the paper channels by a photolithographic technique, similar to that described by Martinez *et al.*[2]. These papers have mean pore diameters of about 11 μm, and have 'medium' porosity (~70-80%). First, the cellulose filter paper is uniformly soaked in a negative photoresist (SU8 10; MicroChem), followed by pre-baking at 110 °C for 10 minutes. The cooled, photoresist-soaked filter paper is then exposed to UV radiation (405nm wavelength) for 20 seconds, through an appropriate photo-mask, by using an UV mask aligner (OAI Hybralign Series 200). The exposed substrate is then post-baked at 110 °C for 10 minutes, and finally the non-cross-linked photoresist is removed by washing with acetone (for detailed fabrication procedure; see ESI, Fig. S1†).

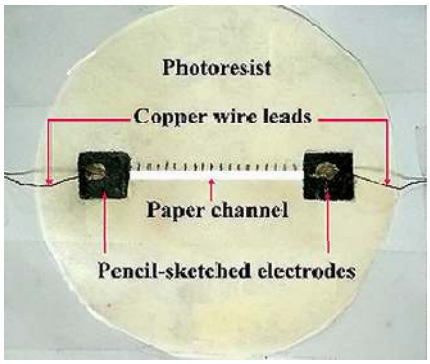

Fig. 1 The 'paper-and-pencil' device for demonstrating the electrokinetic phenomenon in paper substrates.

The fabricated device (see Fig. 1) consists of a 1 mm wide channel, interconnecting two 1 cm×1 cm square pads, which are 4 cm apart. The electrodes are 'sketched' onto these two square pads by using a 2B lead-pencil. Repeated rubbing of the sharpened lead-tip of the pencil, on the paper surface, covers the pads with a thin layer of graphite. Thereafter, thin copper wires (diameter 0.5 mm) are attached to these 'sketched' electrode pads by means of conductive silver paste (Alfa Aesar: sheet resistance < 0.025 ohm/square @ 0.001 inch thick). The copper wires are then connected to the terminals of a DC-power supply (Keithley 2410 1100V sourcemeter), for applying the necessary electric potential. Now, the question that we want to address here is: *do the paper fibres and the electrolyte solution trigger the spontaneous formation of electric double layer[8] (EDL) within the paper-pores, which manifest in controllable electrokinetic flows in the 'paper-and-pencil' device?* To distinctively pin-point the electrokinetic effects, the liquid flow in paper is characterized with and without electrical effects, and thereafter with varying magnitudes of applied electric potential. To further highlight the electrokinetic influence, the paper channel is completely wetted with 1 mM aqueous KCl solution, by normal wicking action, before the electrical actuation. This process fills the inter-fibre passages with electrolyte solution, and tends to reduce further liquid flow due to natural imbibition. Immediately after this,

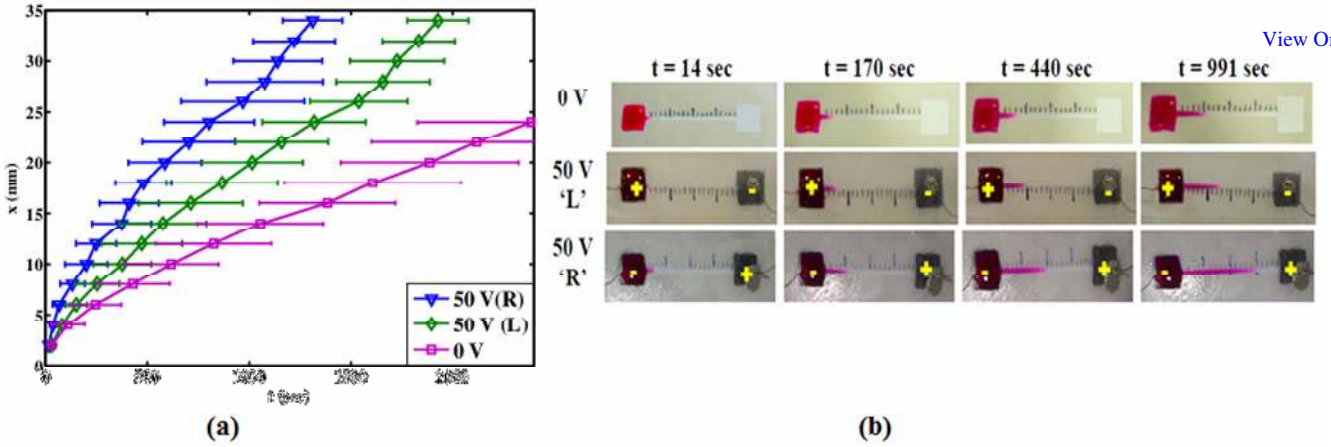

Fig. 2 (a) Variations of liquid flow speeds through the paper channel without electrical effects, and with electrical effects for two different polarities of the applied potential. The solid lines are guide to the readers' eyes only. The errorbars are $\pm\sigma$. (b) Real time images of the advancing liquid front at different temporal instances for pure capillary-driven and electrically aided flows. Comparison of these image sequences reveals the enhancement in liquid transport due to the electrokinetic phenomenon.

80 µl of 1 mM KCl solution, stained with a fluorescent dye (Rhodamine-B; concentration: 0.667 g/l of 1mM aqueous KCl), is manually dispensed on the left electrode pad by a micro-pipette. Then the electric potential is applied across the paper channel, by means of the DC-power supply. Furthermore, the response of the flow dynamics to the change of polarity of the applied potential is revealed by characterizing the flow, in one device, by activating the left electrode pad, i.e. the one containing the liquid reservoir (this configuration is designated here as 'L'); while in another identical device, a similar study, with the same test liquid, is performed by applying the potential to the right pad, relative to the left one (this configuration is designated as 'R'; see ESI, Fig S2†). The flow speed is characterized by specifying the time of arrival of the dyed liquid front at a definite spatial location marker, scribed at equal intervals along the edge of the channel.

The flow speed of the electrolyte, through the paper channel, is distinctly enhanced in presence of an imposed electric potential (50 V), as can be clearly perceived from a comparison of the transport characteristics depicted in Fig. 2a. In the absence of any electrical influence (0 V), the normal imbibition of the dyed liquid, through the pores of an initially dry paper channel, stems from the interplay of capillary and viscous forces, as classically addressed by the Washburn model[9], and diffusion of dye molecules. Now, for the paper-based electrokinetic device, the paper fibers in physical contact with the aqueous KCl solution develop surface charge, through ionization or ion-adsorption, which alters the distribution of the oppositely charged ions (counter-ions) within the electrolyte solution, due to dominating electrostatic attraction. The redistribution of the counter-ions, in the immediate vicinity of the paper fibers, culminates in the formation of electric double layer (EDL). On application of an external electric field, the net free charge, within this spontaneously formed EDL, generates an electrical force. The electrical force 'pulls' the dyed liquid from the reservoir pad, and then through the paper channel, resulting in sustained electroosmotic flow. It is to be noted here that within the paper-pores, electrophoretic motion of the Rhodamine B dyed solution is also triggered[10]. Hence, the electroosmotic flow of the bulk electrolyte, as well as the electrophoretic transport, culminates in faster mass transport through the paper substrate, as compared to that due to classical imbibition. The electrically triggered higher flow speeds in the paper based device is coherently reflected by the differences in spatial distances traversed by the dyed liquid front, over same time intervals, as sequentially exhibited in Fig. 2b. The characteristic of the EDL, which forms the core of the electrokinetic phenomenon, is intrinsically dependent on the electro-surface-chemistry and the pH of the dyed electrolyte solution. Hence, the nature of the free charge density (positive or negative) within the developed EDL, and consequently the direction of the electroosmotic flow through the inter-fiber passages, for a particular polarity of the applied potential, cannot be intuitively ascertained *a priori*, for the cellulose paper-fiber and the aqueous KCl system. Furthermore, the randomly oriented paper-fibers result in tortuous flow passages which impart non-uniform directionality to the local electric field, and hence result in directionally non-uniform electrokinetic liquid transport within the microscopic flow domains. The visually perceived transport of the dyed liquid front through the paper channel of the device is a gross culmination of the existing electro-hydrodynamics within these innumerable paper-pores. Hence, the prediction of a correlation between the direction of observed electrokinetic flow and polarity of the applied potential is difficult. In that respect, for the experiments reported herein, it can be said that although the direction of liquid flow are same, the electrically induced enhancement of the transport phenomenon is relatively greater for the applied polarity in configuration 'R', than that in configuration 'L', for identical location of the liquid reservoir (on the left pad) and magnitude of the applied potential (see Fig. 2).

The inherent irregularity in the arrangement of the paper-fiber matrix can be observed, to a certain extent, in the fluorescence microscopy images (Olympus 1X71) shown in Fig. 3. From these images it can be also unambiguously concluded that the electrokinetically aided liquid transport takes place through the paper structure and not 'over' it, unlike the SAW aided transport in paper based devices[6]. Now, the strength of the electrokinetic transport of the dyed liquid through the paper substrate is proportional to the magnitude of the applied

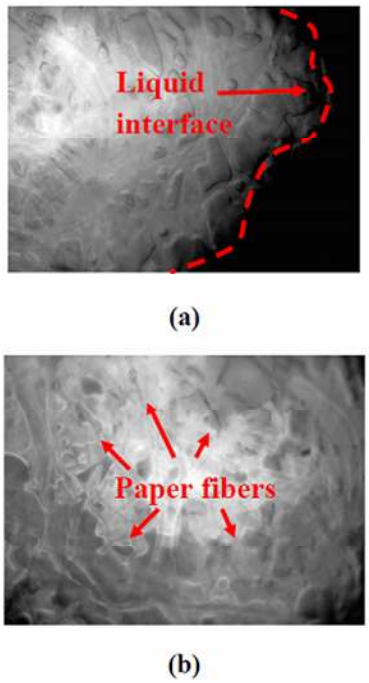

Fig. 3 (a) Fluorescent microscopy image of the dyed liquid front advancing through the paper substrate, under the influence of the electrokinetic phenomenon.**(b)** The liquid flows through tortuous microscopic flow passages formed by the randomly arranged paper fibres.

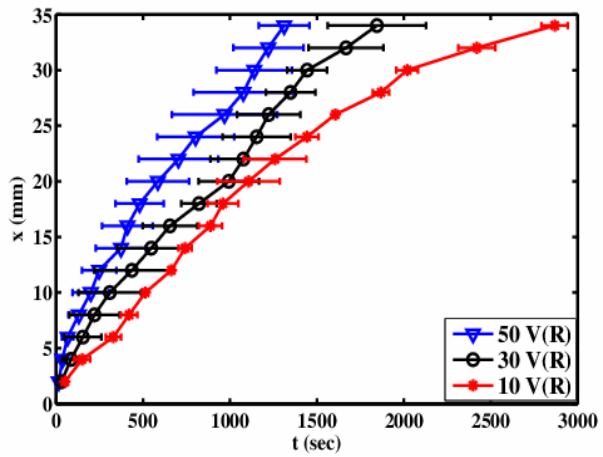

Fig. 4 Variations in liquid flow characteristics through the paper channel, for progressively increasing magnitudes of the applied electrical potential. This clearly reflects the controllability and repeatability of the electrically aided liquid transport in the 'paper-and-pencil' device. The solid lines are guide to the readers' eyes only. The errorbars are $\pm\sigma$.

electric potential, as in general, the aiding electroosmotic force is linearly proportional to the applied potential[11]. This underlying physics is exploited to gain active electrical control over the transport phenomenon in the proposed 'paper-and-pencil' device. The alterations in the liquid flow characteristics, through the paper channel, with variations in the applied electric potential for the more advantageous 'R' configuration, are delineated in Fig. 4. Hence, the electrokinetically induced transport of the liquid in the paper-device not only has improved repeatability, as compared to normal capillary-driven flow (apparent from the involved errorbars in Fig. 2a and Fig. 4), but also has significant controllability.

The electrical control of liquid flow in the 'paper-and-pencil' device can be implemented for significantly improving (both repeatability and controllability) the present modality of immunochromatographic assays. Moreover, the existence of electrokinetic phenomenon in paper substrates, as demonstrated here, can be exploited for performing various processes, for e.g. manipulation/trapping of electrically responsive biomolecules, electrical separation/mixing of analytes, and microscale liquid handling, all of which have been never tried before on inexpensive and easily manufacturable paper devices. Furthermore, the proposed electrical forcing mechanism is very simple, which does not require any additional actuation devices, and hence can be easily integrated even with intricate microfluidic networks on paper substrates without compromising the cost-effectiveness and the portability of such devices. In essence, the utilitarian scope of the "electrokinetics with 'paper-and-pencil' devices" is far-reaching and still open to imagination.

Notes and references

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