Sensors and Actuators B 177 (2013) 620–626

Contents lists available at SciVerse ScienceDirect

Sensors and Actuators B: Chemical

journal homepage: www.elsevier.com/locate/snb

Optimized micro devices for liquid-dielectrophoresis (LDEP) actuation of conductive solutions

R. Renaudot[a,*,1], B. Daunay[b,1], M. Kumemura[b,c], V. Agache[a], L. Jalabert[b], D. Collard[b], H. Fujita[c]

[a] CEA-LETI, Minatec Campus, 38054 Grenoble, France
[b] LIMMS-CNRS/IIS (UMI 2820), The University of Tokyo, 153-8505 Tokyo, Japan
[c] IIS – Institute of Industrial Science, The University of Tokyo, 153-8505 Tokyo, Japan

ARTICLE INFO

Article history:
Received 17 August 2012
Received in revised form
14 November 2012
Accepted 15 November 2012
Available online 27 November 2012

Keywords:
Conductive/dielectric solutions
Liquid dielectrophoresis
Actuation voltage
Droplets generation

ABSTRACT

Liquid dielectrophoresis (LDEP) technique is known to displace liquids in open environments by applying alternating or DC voltages between electrodes patterned below a single or a stack of dielectric layers. This technique is able to handle pico–nano liter droplets and could be used to carry out biological and chemical protocols. According to the electrode geometry, the dielectric layers constants and thicknesses, this technique may require relatively high actuation voltages (~200–500 V_{RMS}). In addition, most of experiments carried out in the literature have shown the LDEP technique to be more adapted to insulating or semi insulating liquids (lower than 1 mS m^{-1}). This paper reports optimization of LDEP devices, in terms of miniaturization (generation of droplets arrays with droplets size ranging from 0.5 pL to 0.1 nL), and choice of dielectric materials. Following this procedure we have successfully addressed two challenging constraints in the field of LDEP: firstly deionized water can be actuated with operating voltages below 150 V_{RMS} and secondly solutions with conductivity up to 10 mS m^{-1} have been partly displaced. These results allow us to foresee manipulation of organic solutions, biological buffers by LDEP at the micro-nanoscale, which will attract strong interest for sample preparation in complex biochemical protocols.

1. Introduction

In the last twenty years, digital microfluidics (DMF) has emerged as a promising versatile tool to manipulate a wide range of liquids on a planar surface using electromechanical forces [1,2]. Among those techniques, liquid dielectrophoresis (LDEP) and electrowetting on dielectric (EWOD) actuations have already been used to address elementary microfluidic operations [3–6] and/or complex biochemical protocols [7–11] with accuracy and reproducibility in terms of liquid displacements. As usually described in the literature, LDEP is an electromechanical response of an insulating liquid, either confined between two opposite electrodes (two-plates configuration) [12–14], or deposited onto two coplanar electrodes (open single-plate configuration) [15–17] separated by a gap and polarized using a non uniform or DC electrical signal. The origin of the LDEP electrical driving method and its relationships with EWOD method have been discussed in-depth by Jones et al. and Wu et al. [18–20]. In the open single-plate configuration, LDEP transport, qualified sometimes as a SMF (surface microfluidic) technology, allows the splitting of a mother droplet (~1 μL) in arrays of smaller ones (from pL to nL) within a very short time (~100 ms). LDEP technique can provide quickly and simultaneously an array of micrometric size controlled droplets (radius in the μm range), which is beneficial for carrying out sample preparation required for massively parallel and complex biochemical protocols (see Fig. 1). Usually, this technique requires actuation voltages which are above 200 V_{RMS} for an open single-plate microfluidic configuration, at frequencies ranging from DC up to the MegaHertz [15–17,21,22].

Moreover, the first studies investigated LDEP actuation with strongly insulating liquids such as oil [23,24] and the technique has progressively been expanded in the time to more conductive liquids and semi-insulating liquids [25]. Generally, the original LDEP microdroplets dispensing were largely restricted to deionized (DI) water. To the best of our knowledge, the maximum reported conductivity is 1 mS m^{-1} so far [26], even if theoretical models predict that conductive solutions could be displaced provided that both the signal frequency and voltages are high enough [27,28]. As previously identified by Chugh et al. [29], in this study we propose to address two main challenges:

1. Reduce actuation voltage by optimizing the dielectric material stacks and their thicknesses (as studied by Saeki et al. [30] or Moon et al. [31] for EWOD actuation) aiming at driving the

* Corresponding author. Tel.: +33 04 38 78 14 43.
E-mail address: raphael.renaudot@cea.fr (R. Renaudot).
[1] These authors contributed equally to this work.

0925-4005/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.snb.2012.11.049

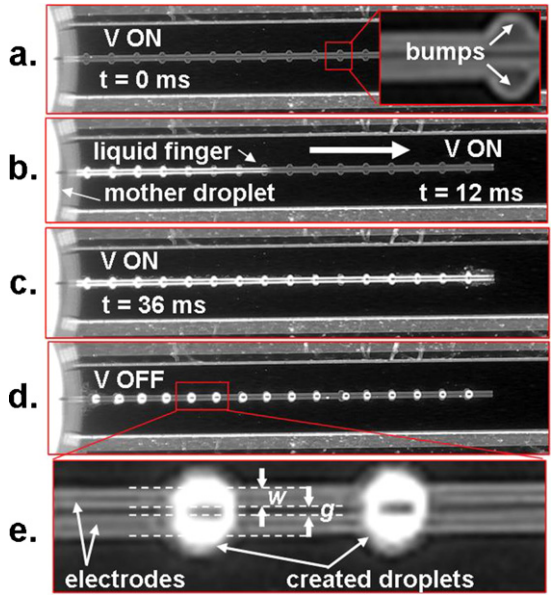

Fig. 1. (a)–(c) Evolution of a DI water liquid finger while applying a 250 ms signal pulse of 236 V_{RMS} at 100 kHz. The dielectric layers are made of 300 nm SiN and 300 nm SiOC (see Section 2.2). (d)–(e) The picoliter droplets are created by the break-up of the capillary instability at semi circular profiles positions called bumps [28]. The electrodes width w is 7 μm while the gap g is 4 μm. This photos sequence is obtained with a high speed camera (250 frames per second).

microfluidic chip electrodes with co-integrated CMOS circuit and standard electronic equipments.

2. Displace higher conductive liquids (>1 mS m^{-1}) which is of most concern for biochemical applications. Indeed, the conductivities of samples such as typical biological buffers (TRIS, PBS. . ., etc), DNA buffers and physiological liquids are generally in the order of magnitude of 1 S m^{-1}. Hence, pumping more conductive liquids by LDEP transport holds a high interest and is a key issue to broaden the range of feasible biochemical protocols. Because the LDEP is based on electrostatic actuation, such high conductive liquid can be theoretically displaced using a high frequency and high voltage signal range.

This paper presents optimized dielectric stacks implemented on LDEP devices that first prevent from dielectric breakdown, and secondly allow actuation of relatively high conductive liquids (up to 10 mS m^{-1}, ten times more than the reported maximum value in the literature [26]) with a reduced actuation voltage (around 100 V_{RMS} for DI water, which can be considered as a reference liquid solution). The optimization of the dielectric stacks has been made according to a theoretical model taking into account mainly the electrical and wetting properties of the top layer of several materials, including dielectric multi-stacks. Then, the generation of several droplets, from a typical conductive DNA buffer (diluted 10 times in DI water) has been demonstrated. Those experimental results have been systematically compared with the theoretical model. Finally, some limitations concerning the top dielectric layer wetting properties and undesired phenomena, such as Joule heating, in actuating higher conductivity liquids are discussed.

2. Materials and methods

2.1. Theoretical model

According to the theory developed by Jones et al. [27,28] and Chatterjee et al. [32,33], the threshold liquid actuation voltage V_{th} depends on several parameters: mainly the liquid properties, the dielectric layers properties and the electrodes geometry (see Eqs. (1)–(5)). In this study, the chip is modeled as a stack of N dielectric layers, where each layer i can be made of silicon nitride (SiN), hydrogenated silicon oxycarbide (SiOC) film, or Teflon layer (TFL). As detailed in Renaudot et al. [34], the actuation threshold voltage V_{th} is given by:

$$V_{th} = \sqrt{\frac{2F_\gamma \left(\sum_i^N (2/C_i^*) + (1/C_{liq}^*)\right)\left(\sum_i^N (2/C_i^*) + (1/C_{air}^*)\right)}{\left((1/C_{air}^*) - (1/C_{liq}^*)\right)}} \quad (1)$$

$$V_{th} = \sqrt{\frac{2F_\gamma\{\alpha^2(2/\varepsilon_0 w)^2 + \alpha(2/\varepsilon_0 w)((1/C_{air}^*) + (1/C_{liq}^*)) + ((1/C_{air}^*) + (1/C_{liq}^*))\}}{(1/C_{air}^*) - (1/C_{liq}^*)}} \quad (2)$$

$$C_i^* = \frac{\varepsilon_0 \varepsilon_i w}{d_i} \quad (3)$$

$$\alpha = \sum_{i=1}^{N} \frac{d_i}{\varepsilon_i} \quad (4)$$

$$F_\gamma = \pi \left(w + \frac{g}{2}\right) \gamma_{liq} \quad (5)$$

In expressions (1)–(5), w, g, d_i, ε_0, ε_i relate respectively to the electrodes width, the inter-electrode gap, the dielectric layer i thickness, the vacuum permittivity and the dielectric constant of the dielectric layer i. C_i^*, C_{liq}^*, C_{air}^* represent the electrical capacitance of the dielectric layer i, the liquid and the air derived with respect to the direction of liquid motion. F_γ is the force generated by the liquid surface tension in the air, γ_{liq} being the liquid surface tension in the air. In expression (4), α is defined as the equivalent dielectric thickness of a layers stack and strongly affects the V_{th} value. Recently demonstrated in Daunay et al. [35], the DI contact angle of the liquid sample on the surface influences the liquid actuation. A new expression of V_{th}, called $V_{th/\theta}$ is then established with a corrective factor depending on the contact angle θ:

$$V_{th/\theta} \approx \frac{2\theta}{\pi} V_{th} \quad (6)$$

In addition to V_{th}, a second parameter is usually considered: the threshold actuation frequency f_c defined as following [33]:

$$f_c = \frac{1}{2\pi}\frac{g_{liq}}{C_{eq}} = \frac{1}{2\pi}\frac{g_{liq}^*}{C_{liq}^* + 1/2 \cdot \left(\sum_{i=1}^N 1/C_i^*\right)} \quad (7)$$

In expression (7), g_{liq} stands for the liquid conductance derived with respect to the liquid displacement direction. Consequently, for a given device design and material arrangement, the higher the liquid conductivity is, the higher the LDEP threshold actuation frequency is.

2.2. Chip configuration and fabrication

Open single-plate architecture devices, as shown in Fig. 2a, have been fabricated with driving electrodes widths w ranging from 3 to 20 μm and inter-electrode gaps g ranging from 2 to 10 μm. Considering such designs, created droplet volumes should theoretically range from 0.5 pL to 0.1 nL [36], while the initial mother droplet volume is about 1 μL. The driving electrodes are made of Ti (10 nm)/AlCu (200 nm), deposited by electron beam evaporation. The thin Ti layer plays the role of adhesive layer for a well controlled photolithography and etching process. The AlCu layer is a mix of majority Al metal and Cu metal, commonly used in microelectronics process. Electrodes are patterned by standard photolithography and etched on a silicon substrate (750 μm thickness) covered by a