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INVITED REVIEW PAPER

Next generation digital microfluidic technology: Electrophoresis of charged droplets

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Abstract−Contact charging of a conducting droplet in a dielectric medium is introduced as a novel and useful digital microfluidic technology as well as an interesting scientific phenomenon. The history of this phenomenon, starting from original observations to its interpretations and applications, is presented. The basic principle of the droplet contact charging is also presented. Several fundamental aspects of the droplet contact charging from view points of electrochemistry, surface science, electrocoalescence, and electrohydrodynamics are mentioned. Some promising results for future applications and potential features as a next generation digital microfluidic technology are discussed, especially for 3D organ printing. Finally, implications and significance of the proposed technology for chemical engineering community are discussed.

Keywords: Microfluidics, Droplet, Contact Charging, Electrophoresis, Charge Transfer

INTRODUCTION

When a water droplet suspended in a dielectric medium is applied to a strong electric field, the water droplet bounces back and forth between the two electrified electrodes as shown in Fig. 1 [1-5]. Interestingly, even though we apply just a static electric field (the field direction does not change with time), the water droplet keeps bouncing. In spite of the direct contact with electrode surface, because the contact area is so small that the water droplet does not leave any remnant on the electrode surface. Because we need a somewhat high electric field, the application area of this phenomenon seems to be quite limited. However, when the system size is on a micrometer scale, the applied voltage can be much reduced to several tens of volts [2,6]. Therefore, this droplet contact charging phenomenon on an electrified electrode has great potential for digital microfluidic technology which uses droplets as tiny compartments inside lab-on-a-chip devices.

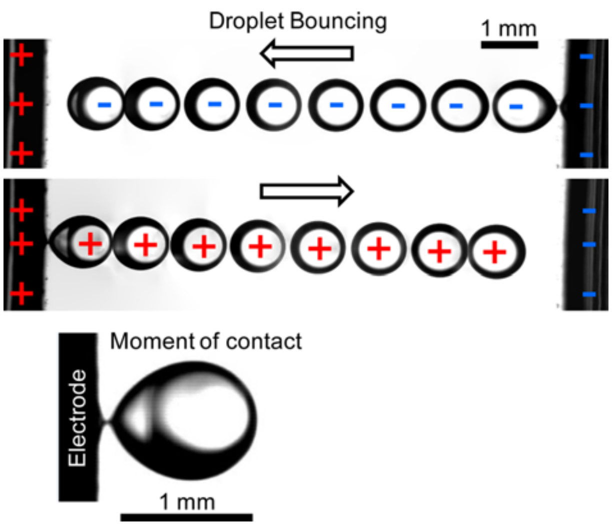

Fig. 1. Bouncing motion of a 300 nL water droplet immerged in dielectric oil (silicone oil) due to the contact charging phenomenon. Applied electric field strength is 3 kV/cm. The time step of time-lapse image is 0.06 sec.

By controlling an individual droplet movement using electric field, we can perform bio and chemical processes in a micrometer scale. Electrowetting and dielectrophoresis are the two major technologies in this field [4,7]. Electrowetting or Electrowetting on dielectric (EWOD) method is also referred to as digital microfluidics (DMF) because droplets are manipulated on the array of electrodes [8-10], and this characteristic is a good match for the array-based biological and chemical applications [11,12]. But electrowetting requires a droplet to be in contact with a surface, which causes contact line pinning and bio-fouling due to surface contamination [7]. In dielectrophoresis (DEP), a droplet does not have to contact a surface [13-15]. The DEP force depends only on the dielectric property of droplet, which provides the capacity for processing droplets composed of various media and promises great versatility across a wide spectrum of applications [14,16-18]. However, the droplet actuation method using DEP is not popular, as EWOD because it needs higher voltage [19] and the resulting droplet velocity is slower than EWOD method [14].

The droplet contact charging phenomenon, referred to as 'electrophoresis of a charged droplet (ECD) [4]' or 'contact charge electrophoresis (CCEP) [20]', has many advantageous features as a new digital microfluidic technology [4,21-23]. First, ECD is free from surface contamination. Although there is direct contact with electrode surface when charging occurs, the contact area is very small as shown in Fig. 1, and the contact time is less than a millisecond

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‡This article is dedicated to Prof. Hwayong Kim on the occasion of his retirement from Seoul National Univerisity.

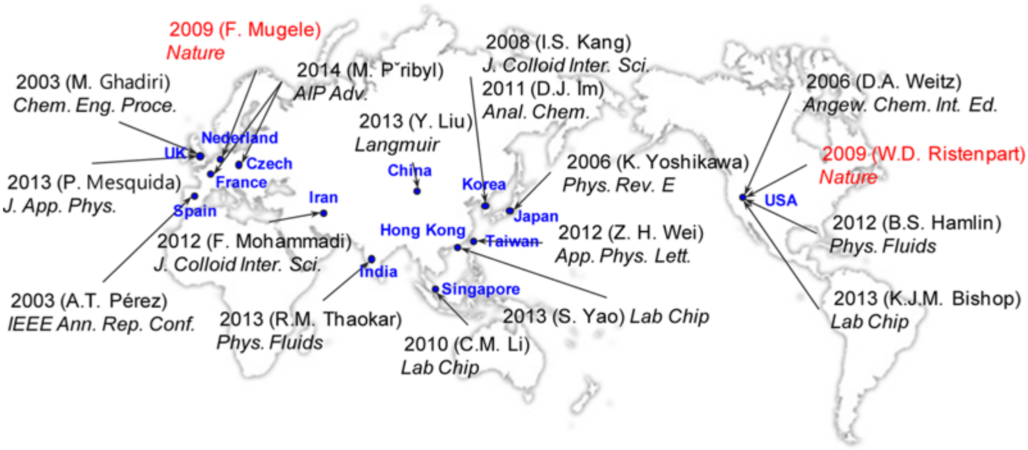
[24]. Second, electrophoresis is simple and straightforward in principle, which provides high degree of freedom in chip design. Because we should understand complicated variation of surface tension and induced many theoretical [25-27] and numerical [28,29] researches to explain the underlying physics. However, the electrophoretic motion of a charged droplet can be easily predicted once the amount of charge of the droplet is known under the given electric field. Third, the coalescence of droplets can be easily controlled. It is difficult to merge droplets in a microchannel, especially when the droplets are stabilized with surfactant, which has limited the widespread use of droplet technology as robust micro reactors in microfluidic devices [30,31]. An electric field can be used to make charged droplets to merge in a microchannel [30-32] quite easily, and furthermore, under sufficiently high electric field, we can also make droplets not to merge [33,34].

In addition to the above mentioned digital microfluidic applications, the droplet contact charging phenomenon can also be used in other application areas. In microchannels, the droplet charging phenomenon is utilized as the coalescence [35,36], sorting [37,38], trapping [39], and fission [35] of droplets with carrier fluid. Electrocoalescence and breakups of drops are an important research subject in climate science [40,41] as well as in the dehydration process of crude oil [42,43]. Liquid contact electrification charging of drops can be employed in portable energy harvesting devices [44-50]. Understanding of the charged droplet behavior under electric field is critical in electrospray mass spectrometry [51-54] and electrohydrodynamic (EHD) printing technology [55,56]. In physical chemistry, the charging phenomenon can also be used in measuring interfacial tension of ionic liquid/silicone oil system [57] and in extracting specific ions from ionic liquid [58].

Here in this review, a novel digital microfluidic technology based on the droplet contact charging phenomenon is introduced because it has great potential for bio and energy related applications. To give a complete review for this field, not only related applications but also fundamentals are discussed. We will start with the history of this interesting finding from the beginning to the recent works. Next, the basic principle of the droplet contact charging phenomenon will be explained. In the following fundamentals and applications section, important research works will be covered in each category. Finally, the future outlooks of the proposed technology will be discussed.

HISTORY

A charged droplet can electrophoretically move [59]. Millikan measured the elementary charge using charged oil drops in air in his famous classic measurements [60]. Charges at the water-hydrophobic medium interface have been the subject of considerable



Continent	Research Group	Research Focus and Novelty
Europe	M. Ghadiri	Electro-coalescence, Dehydration of crude oil
	F. Mugele	Electro-coalescence, Surface Science, Dehydration of crude oil
	P. Mesquida	Contact Charging Phenomena, Surface Science
	M. P˘ribyl	Contact Charging Phenomena in a microchannel
Asia	K. Yoshikawa	Drop dynamics under contact charging electrophoresis
	I. S. Kang and D. J. Im	Contact Charging Phenomena, ECD based Digital Microfluidics
	R. M. Thaokar	Electrohydrodynamics under non-uniform electric field
	Y. Liu	Contact Charging Phenomena, capacitive measurement of charge
	S. Yao	Microchannel applications, Non-contact electrostatic charging
America	D. A. Weitz	Microchannel applications, splitting, sorting unit development
	W. D. Ristenpart	Electro-coalescence, Electrohydrodynamics, stagnant cap model
	K. J. M. Bishop	Contact Charge Electrophoresis(CCEP) of particles

Fig. 2. Research groups all over the globe reporting the droplet contact charging phenomenon.

scientific interest and debate, due to their significance for emulsion stability and in various applications in the field of colloid science [61-65] and microfluidics [66]. Storm cloud formation can be explained by the coalescence of charged water drops in free fall [40].

Although the above mentioned works are somewhat related to the droplet contact charging phenomenon, more directly related original work of this field can be a report by Mochizuki et al. [67]. They reported translational motions of liquid drops in an immiscible, dielectric liquid confined by a pair of tilted parallel plate electrodes, across which a steady electric field is being applied. They focused on heat transfer enhancement and solvent extraction by the droplet motion. Interestingly, for a decade, no related research was reported after their work. In early 2000s, two research groups worked on this subject again. Eow et al. reported a series of work for the charging of water drops in oil focusing on dehydration of water from crude oil [1,42,43]. Khayari and Perez reported a bouncing motion of a spherical metallic ball between two parallel plate electrodes by measuring and analyzing the charge amount acquired by the bouncing ball [68]. However, none of these works was directly related to microfluidics. In 2006, two papers were published in microfluidics field. Link et al. reported electric control of droplets in microchannel [69] and Hase et al. reported the dynamics of a small droplet between pin shaped electrodes pair [2]. From 2008, Jung et al. reported a series of work related to a digital microfluidic technology without using carrier fluid [3,70,71].

In 2009, two papers were reported in the journal *Nature* directly related to this subject [34,41]. In addition to their work, in 2009 only, three more papers were published in the subject of electro-coalescence [31], breaking of an emulsion under electric field [32] and digital microfluidics [71]. From this period, all over the globe, increasing numbers of work have been actively reported in this field as shown in Fig. 2. Fundamentals of electro-coalescence and breakups of drops [70,72,73] is one of the most closely related research topics. In Physical chemistry, the droplet contact charging phenomena was used in understanding intermolecular forces between two immiscible fluids system by measuring the interfacial tension [57] or by molecular dynamics simulation [66]. Modeling of drop/particle motion experiencing the contact charging phenomenon is one of the key subjects in this field [4-6,66,74-82]. In microfluidics field, the direct contact charging is used to manipulate droplet [38,83-86] or particle motions [20,75] in microchannels in addition to digital microfluidic approaches [6,23,70,77,87-89]. The details of each category will be covered in the following fundamentals and applications section.

BASIC PRINCIPLE

Droplet contact charging phenomenon can occur with or without application of external electric field [50]. However, usually, the ECD or CCEP refers to a contact charging phenomenon using applied electric field. Then, why and how does a droplet acquire charges? When a conductive sphere is brought in contact with an electrified electrode, the sphere is charged as a result of redistribution of electric field around the sphere as shown in Fig. 3. According to perfect conductor theory, the charge amount that the droplet acquires on an electrified electrode Q is proportional to the surface area of the sphere and applied electric field [4,68,90,91].

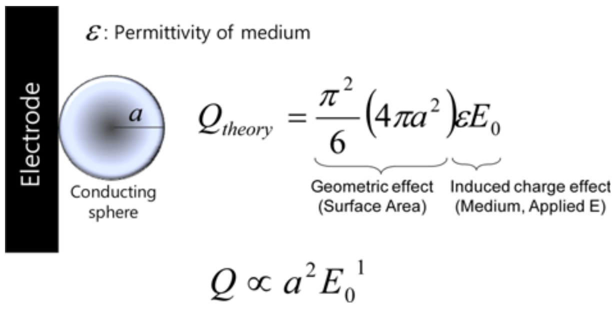


Fig. 3. Perfect conductor theory: charging of a perfectly conductive sphere on an electrified electrode. E_0 is applied electric field strength.

Even though an aqueous droplet in dielectric medium might be considered as conductor, there are still some points to be considered before adopting the perfect conductor theory. First, an aqueous droplet has deformable surface. The deformation of a water droplet upon contact with electrode can affect the charging [80]. Second, water has finite conductivity. Because the charging occurs in less than a millisecond, relatively low conductivity may limit the charging amount to a value that is smaller than the theoretical maximum. Third, unlike metallic conductor, the charge transfer by free electron movement may not be possible in water. It seems to have different charge transfer mechanism such as electrochemical reaction of ions [4]. Therefore, to address the above mentioned concerns, there have been a number of researches. Eow and Ghadiri [1] reported the droplet bouncing motion, but they focused more on the deformation and breakup rather than on the droplet motion and charge amount. Hase et al. [2] focused on the dynamics of droplet motion rather than on the comparison between perfect conductor theory and experimental observations. Khayari and Perez [68] and Jung et al. [3] tried to compare the experimental observations with perfect conductor theory but there was a discrepancy between theory and experiments.

One of the difficulties of droplet contact charging experiment is that it is very sensitive to small dust inside dielectric oil and water phase. In addition, deformation of water droplet also makes the comparison between theory and experiment difficult. Therefore, a specially designed experimental setup was devised and a comparison was successfully performed as shown in Fig. 4 [78]. To obtain reproducible experimental results, a cuvette was used as a disposable oil container together with a special electrodes pair for clean experiment. To minimize deformation effects, the range of droplet size and applied electric field was carefully selected. By measuring the droplet charge using an electrometer and hydrodynamic analysis, it was found that, basically, the droplet contact charging phenomenon can be explained by the perfect conductor theory [78].

1. Fundamentals and Applications

From an electrochemical point of view, ECD motion has a unique feature as shown in Fig. 5(a). In conventional electrochemical systems, positive and negative electrodes are connected through electrolyte solution to transfer charges. However, in an ECD system,

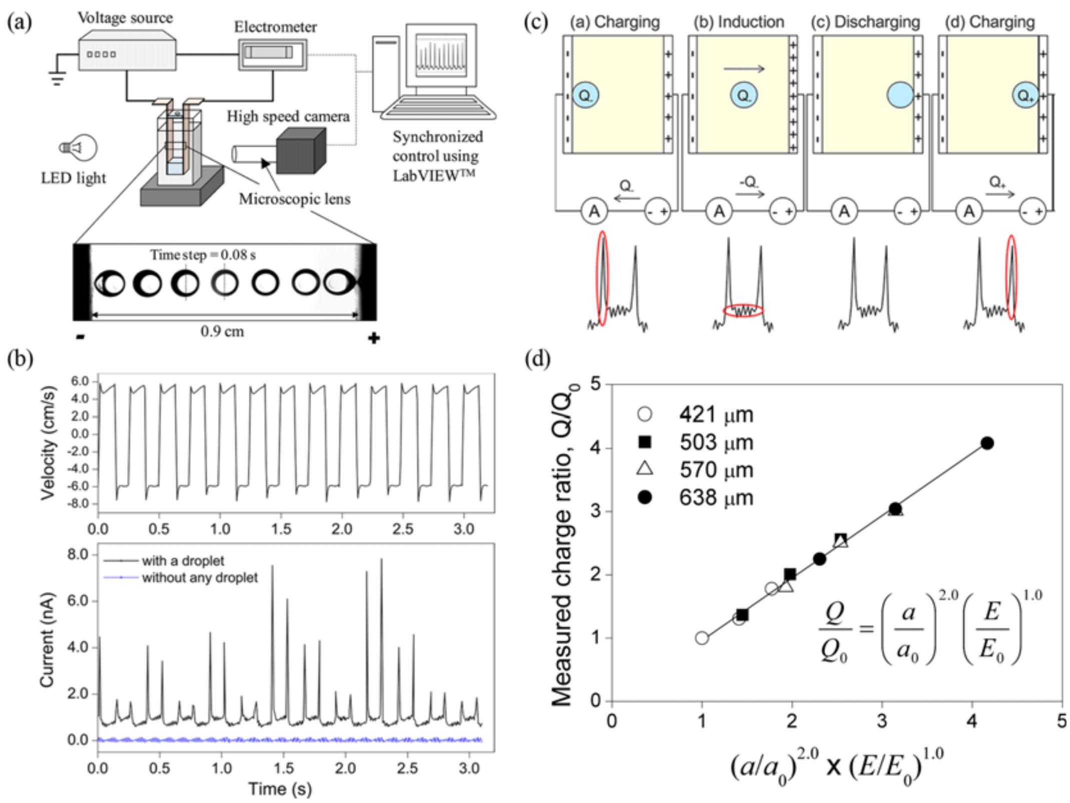


Fig. 4. The charge transfer mechanism of droplet contact charging phenomenon. Measurement of droplet charge and comparison with perfect conductor theory. (a) Experimental setup. (b) Droplet velocity and current signal for droplet bouncing motion. (c) Charge transfer mechanism by the ECD bouncing motion. (d) Scaling law for the charges measured by an electrometer. Adapted from ref. [78]. Copyright 2012 American Chemical Society.

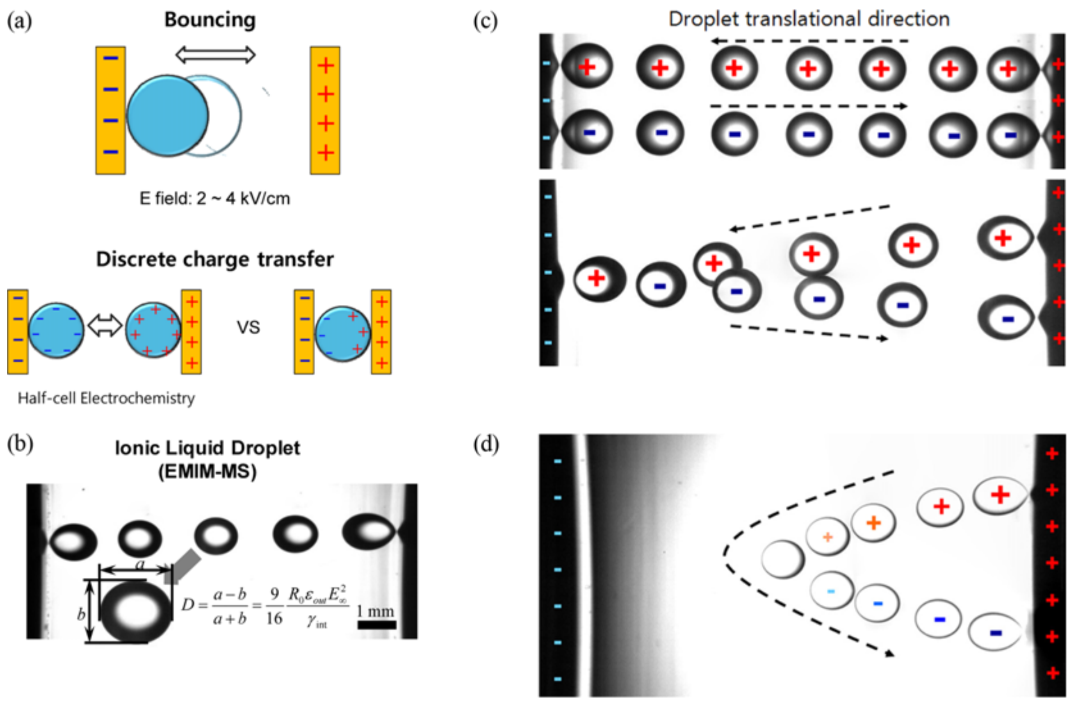


Fig. 5. (a) Discrete charge transfer by ECD. (b) Interfacial tension measurement by ECD. (c) ECD motion of a 300 nL water droplet (top) and a 400 nL BMIM-MS ionic liquid droplet (bottom). (d) Retreating behavior of a 400 nL EMIM-NTf2 ionic liquid droplet. Adapted from ref. [57-58]. Copyright 2013, 2014 American Chemical Society.

charges can be transferred without direct connection between positive and negative electrodes. Therefore, the observation on the separate charging (positive and negative) is possible: the charge is transferred discretely by the movement of a droplet in a dielectric liquid, and we can separately monitor each charge transfer. This unique feature can be utilized to study the charge transfer of cationic and anionic species separately [78]. In physical chemistry, the ECD system can be utilized for understanding intermolecular forces by theoretically [92] and experimentally [57,58] investigating interactions between two immiscible fluids system (ionic liquid/silicone oil) or by molecular dynamics simulations [66]. In surface science, controlling the surface charge on a droplet by chemical or physical methods is an interesting topic [93,94]. Besides, the droplet charging by liquid-solid contact electrification has been actively reported as a novel energy harvesting method [44-48,95].

In physics, droplet behavior under electric field is one of the major concerns of classical electrohydrodynamics [27,96-98]. Especially, electrocoalescence and breakups of drops have many implications in basic science and engineering fields [34,40,41,43,72]. The non-coalescence phenomenon of oppositely charged drops in strong electric fields is a widely-acknowledged fluid dynamics problem [34,41]. In industrial applications, the electrically-induced droplet motion can be used in the dehydration of petroleum and vegetable oil [42,43]. Although non-electrical drop dispensing method is more widely used [99], electrohydrodynamic inkjet printing is also important as an alternative dispensing technology, where a better understanding of the behavior of charged droplets is crucial for accurate control [55,100]. Furthermore, in electrospray mass spectrometry, understanding and control of the charged droplet motion under high electric field is the key [51-53].

To use droplets as tiny compartments inside microfluidic lab-on-a-chip devices, it is important to have information about the electrophoretic behavior and charge of droplets [66]. In that sense, modeling of droplet motion is one of fundamental subjects of ECD. In classical literature, electrophoretic behavior of charged drop were studied theoretically [101] and experimentally [102]. Some early experimental works focused on the dynamics of charged droplet motion [2,103] or on electrohydrodynamic analysis [104] but did not concern the droplet charge measurement. Eow and Ghadiri started to estimate the charge by measuring droplet velocity using time-laps images [1]. Jung et al. also tried a similar approach based on Stokes' regime assumption [3]. Those researches qualitatively explained the phenomenon by comparison with theory but it was not so successful from a quantitative point of view. To address those discrepancies, various approaches had been tried. Khorshidi et al. considered drop deformation effects [80] and Hamlin and Ristenpart used stagnant cap model to explain the hydrodynamic drag force on a moving charged droplet [79]. Measurement of charge using an electrometer can help to elucidate the issue. Khayari and Perez measured charge transfer of a conducting solid particle experiencing CCEP between two parallel planar electrodes pair [68]. Im et al. started the comparison between the results from image analysis and from an electrometer measurement [4,78], and similar approaches were performed by other research groups [74,77]. Recently, ECD under non-uniform electric field by pin-planar type electrodes pair was analyzed [82,88], and modeling of droplet motion in a microchannel setup was also tried [105].

First microfluidic application based on ECD was reported by Link et al. [69], who demonstrated a generic platform technology for manipulating and controlling individual droplets in microchannel. They developed modules that create, recombine, split, and sort droplets one by one, thus providing fine control over individual droplet microreactors. After their work, a number of microfluidic applications in microchannel have been reported. Nui et al. focused on electro-coalescence of droplets [31], Guo et al. [38] and Ahn et al. [84] developed droplet sorting technique for biological applications. Wang et al. developed On-demand droplet release for droplet-based microfluidic system [85]. Rezai et al. reported electrical sorting of a multi-cellular organism such as Caenorhabditis elegans [83]. Zhou and Yao demonstrated non-contact electrostatic charging of droplets by polarizing a neutral droplet and splitting it into two oppositely charged daughter droplets in a T-junction microchannel [86]. Recently, Bishop et al. reported a series of works for contact charge electrophoresis (CCEP) of particles [20,75,106,107]. They focused on the rapid transport of particles or mixing by particle motion in microchannels.

A digital microfluidic approach was also started in late 2000. Jung et al. first reported a digital microfluidic system which can manipulate individual droplet by changing the polarity of electrodes array [70,71]. They showed some potentials for the ECD as a digital microfluidic actuation method; however, in their work, the system size was too large and the actuation voltage was too high (few kilovolts), which limited the applicability of the technology. For that reason, there is a preconception that a high actuation voltage on the order of kilovolts is required for ECD actuation [87], which is not the case for smaller systems [6]. Later, Im et al. resolved this issue by decreasing system size and demonstrated various biochemical applications such as hydrogel and crystal formation as shown in Fig. 6 [23]. Because any conducting droplet can be handled by the ECD method as shown in Fig. 6(a) (even non-aqueous droplets such as glycerol and polyethylene glycol as well as aqueous droplets of milk, coffee, vinegar, and cell suspension), the technology can be applied in various fields [4]. Especially, for biological applications, the viability of cells inside a droplet experiencing ECD actuation is a critical issue because of high electric field application. Fortunately, the viability issue was confirmed for human cells [77] as well as for mammalian cells [89]. The ECD method was also adopted in other digital microfluidic field such as pre-charging method in electrowetting on dielectrics (EWOD) applications [87].

2. Perspectives

Up to now, there is no droplet dispensing technology specifically designed for the ECD based digital microfluidic system. One strong point of EWOD is that dispensing of daughter droplets from a mother drop is well developed (although it is possible for sandwiched type EWOD platform only), which is one of essential parts of digital microfluidic technology. Fortunately, there are some available dispensing mechanisms which can be applied to the ECD system such as electrospray [56] or electric charge concentration (ECC) method [108]. If a dispensing system well fitted to the ECD technology is developed, the applicability of the technology will be much promoted. Another important technique which should be developed for the ECD system is droplet position detection and automatic

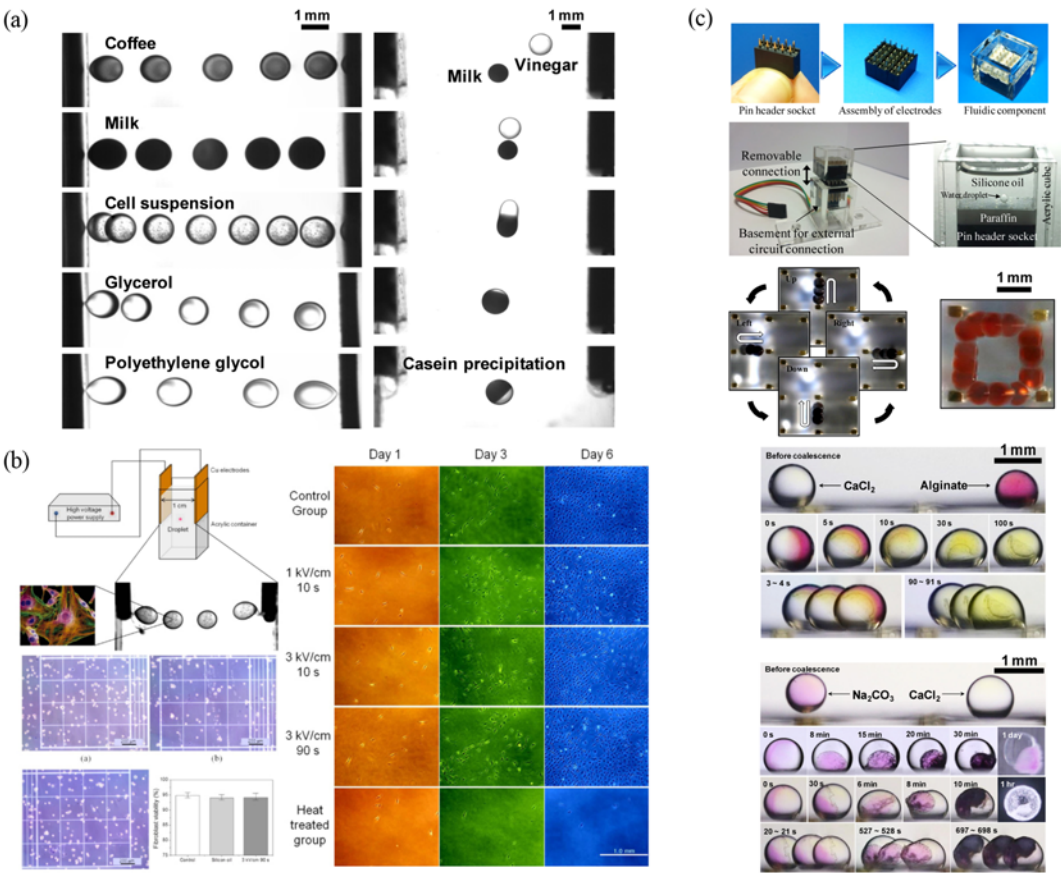


Fig. 6. Digital microfluidic approaches of ECD technology. (a) ECD actuation of a droplet of various aqueous and non-aqueous solutions. Adopted from ref. [4]. Copyright 2011 American Chemical Society. (b) Viability test for mammalian cells (fibroblast). Adapted from ref. [89]. Copyright 2011 American Institute of Physics. (c) Digital microfluidic demonstration of biochemical reactions. Copyright 2013 American Chemical Society.

droplet motion control. Because a droplet under ECD actuation always has certain amount of charges on its surface, this charge can be used to detect the droplet position and therefore can be used for automatic droplet motion control by feedback control of electric circuit [78].

The present ECD-based water-in-oil droplet cell handling approach has great potential for future cell culture applications. Recently, a 3D cell culture technique has received much attention because it is one step closer to an in vivo cell growth environment. However, because 2D culture is easy to carry out and there are no satisfactory 3D cell culture devices available, 2D cell culture is still the predominately used cell culture technique [109]. To resolve the convenience issue, culturing cells inside a small hanging droplet has been proposed as a new approach for a 3D cell culture method [110,111]. Similarly, the water-in-oil droplet adopted in the ECD technique also provides a 3D cell culture environment without any gas exchange problem [112]. Furthermore, by gathering numerous droplets of cell suspension, it is also possible to form an artificial tissue which can be utilized in 3D organ printing [113].

CONCLUDING REMARKS

Droplet contact charging and subsequent electrophoretic motion under electric field (here we called this as ECD or CCEP) was introduced as a novel and useful digital microfluidic technology as well as an interesting scientific phenomenon. We reviewed the history of this phenomenon and also discussed on the underlying basic principle of this phenomenon. Up to now, it has been believed that the droplet contact charging can be explained by classical electrostatics theory even though more detailed studies on the charge transfer mechanism need to be explored. We also reviewed several fundamental scientific aspects of droplet contact charging from view points of electrochemistry, surface science, electrocoalescence, and electrohydrodynamics. Some promising results for future applications were introduced and potential features as a next generation digital microfluidic technology were also discussed, especially for 3D cell culture and organ printing. Considering the impacts of the ECD technology for future bioengineering and biomedical fields, more active participations from various fields are needed to exploit the advantages of the present technology. The hope for the near future is to see a more useful cell culture and engineering platform based on the proposed ECD technology.

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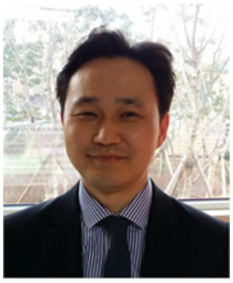

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