Study and Stabilization of a Liquid Crystal Drop Formation Using a Piezoelectric Inkjet Printhead

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Abstract

The development of inkjet printing processes is in great expansion for the fabrication of Microsystems thanks to less material waste and accurate dot placement. The quality of drop formation is known to depend on two main parameters: fluid viscosity and piezoactuator applied voltage. We are working with the thermotropic liquid crystal E7 which presents a nematic oriented phase for a temperature under 60°C and an isotropic phase above 60°C. Like other liquids, viscosity of thermotropic liquid crystals decreases with the temperature but presents a discontinuity at the phase transition, which can disturb the stability of drop ejection. Unfortunately, for the E7 liquid crystal, the piezoprinthead viscosity specifications (usually under 15cP) require a temperature increase close to the temperature transition. Thus, understanding the influence of phase transition on drop characteristics is crucial. Moreover, the applied voltage can be responsible for an electrical field perpendicular to the liquid movements, which can trouble the orientation of nematic phase molecules, change its apparent viscosity and alter the ejection process. In order to understand these phenomena, the influences of viscosity and applied voltage will be studied and discussed. An adjustment tool will be finally proposed to fit with experimental results and to reach finally stable formation of liquid crystal drops.

Introduction

Liquid crystals material exhibit particular phases of matter, called mesophases, between liquid and solid classical phases of conventional liquids. In those mesophases, liquid crystal (LC) inhabit an anisotropic oriented structure, less organized than in the solid state but still have an ability to flow like in its liquid phase. This anisotropy confers to them specific properties, as optical birefringence and high sensitivity to an applied electric field. The study of these properties has received a lot of attention in recent years to create, for instance, liquid crystal wavefront modulators [1] and adaptive lenses [2][3]. Other LC applications have been developed by microelectronic researchers such as tunable capacitors [4] or displays [5].

LC are quite expensive materials, thus the integration process to microsystems has to be very efficient to limit waste of material. Inkjet printing technology, which is in great expansion nowadays in microelectronic fields [6], is a good solution of integration thanks to its very accurate non-contact deposition process of small amounts of material. This is the reason why we are interested in studying and understanding liquid crystal behavior during the drop formation process.

The most commonly used inkjet printing technology is piezoelectric Drop-on-Demand (DOD) printheads. The quality of drop formation in those printheads is known to depend on two main parameters: fluid viscosity (depending on temperature) and driving voltage. Inkjet users have to study these parameters to control volume and velocity of droplets, and ensure an optimized and stabilized material deposition.

Here, we are working with a well known thermotropic liquid crystal, called E7, discovered in 1974 by Raynes [5]. Thermotropic liquid crystals exhibit a phase transition from mesophases to liquid phase as temperature is changed. These liquids present complex rheological properties, owing to their more or less organized states. E7 presents only one LC phase, which is a nematic phase, where molecules are oriented in the same direction but have no positional organization.

In this paper, we will first present XAAR piezoprintheads used, E7 liquid crystal properties and tools used to characterize rheological properties and drop formation process. Then temperature dependency of E7 viscosity in the nematic and the isotropic phase will be detailed. The study of its drop formation process under the influences of driving voltage and viscosity will be presented and discussions on these results will be proposed. Finally, we will show that understanding how inkjet parameters influence drop properties enables to have an optimised and stabilized ejection of liquid crystal drops.

Experiments

In order to understand the behavior of liquid crystal in inkjet printing technology, we have studied the drop formation process of a thermotropic liquid crystal in a XAAR318® printhead. These inkjet printheads are drop-on-demand printheads. They use the deformation of piezoelectric actuators to create a volume change in the channel, which generates a pressure wave that propagates across the liquid at a high shear rate. This acoustic pressure wave overcomes the viscous pressure loss in the small nozzle aperture and surface tension force of the meniscus so that a drop can be ejected from the nozzle. These multi-nozzle printheads can work in greyscale mode [7], deposition of variable size drops of liquid, from 6 to 42 pl, at a frequency comprised between 1 and 4 kHz. Here we have limited the observations to the 6pl drop formation. The viscosity of liquid to be ejected has to be in the range of 6-15cP for a temperature lower than 65°C, and a surface tension from 20 to 40 mN/m.

Considering these requirements, we have chosen to work with E7 from Merck, a four-component thermotropic liquid crystal mixture of cyanobiphenyls, and a cyanoterphenyl, containing 51% of 5CB, 25% of 7CB, 16% 80CB and 8% of 5CT (Figure 1). E7 exhibits a nematic isotropic transition at TNI = 60°C, an adequate viscosity (measured with cone and plate Brookfield viscometer) and a surface tension of 33,9 mN/m at 25°C (measured with Wilhelmy Plate method on a tensiometer from Krüss). These properties enable us to study inkjet drop formation of E7 in its nematic and isotropic phases and during the phase transition. E7 has a positive dielectric anisotropy, owing to a permanent dipole along its molecular long axis. Thus the molecules will orient along the electric field applied.

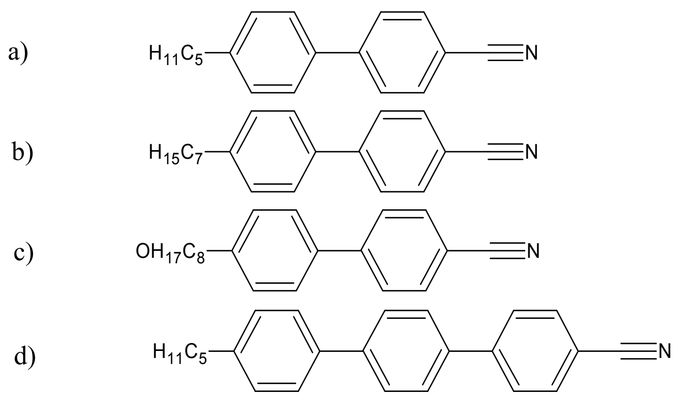

Figure 1 - E7 chemical composition - a) 51% 4-n-pentyl-4'-cyanobiphenyl (5CB), b) 25% 4-n-heptyl-4'-cyanobiphenyl (7CB), c) 16% 4-n-octyloxy-4'-cyanobiphenyl (80CB), d) 8% 4-n-pentyl-4'-cyanoterphenyl (5CT)

The droplets formation has been visualized with a stroboscopic optical system composed of a CCD camera synchronized with a strobe LED system and the printhead. Velocity of ejected drops is measured, while the temperature of the liquid crystal and driving voltage is gradually modified to study their influence. To calculate the velocity, two pictures are taken at two different times; knowing the delay between both pictures and measuring the distance, velocity can be estimated. The frequency of ejection has been set up to 1000 Hz for all the tests presented after.

Results and discussions

First the viscosity variations of E7 versus temperature have been measured (Figure 2). The curve presents a discontinuity at the transition between nematic and isotropic phases. Indeed at TNI, the liquid crystal looses completely his organisation and molecules interactions are suddenly modified.

Far from the transition, both phases are acting like other classical liquids: the viscosity decreases gradually when temperature increases. Then, E7 behaviour can be decomposed in two rheological models: one for its nematic phase and one for its isotropic phase.

The viscosity dependence with temperature of both rheological models follows a classical Arrhenius law:

-Nematic model:

$$\eta = 4.10^{-5}\ e^{4060/(\theta+273,15)} \quad (1)$$

-Isotropic model:

$$\eta = 7.10^{-3}\ e^{2530/(\theta+273,15)} \quad (2)$$

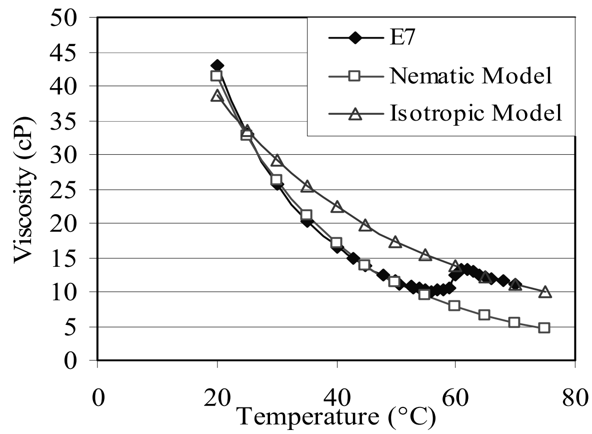

Figure 2 – Viscosity versus temperature of E7 and rheological nematic and isotropic models

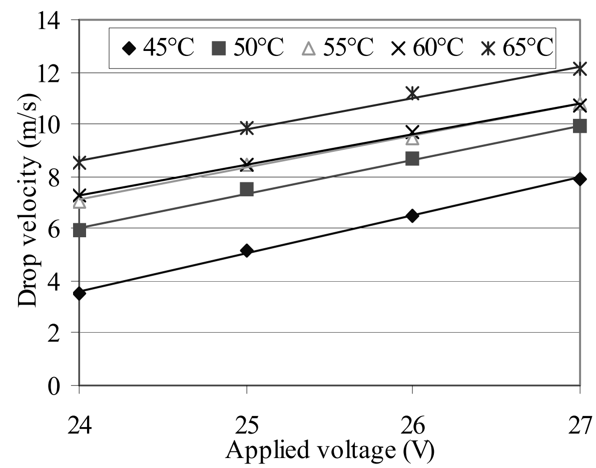

Figure 3 –Drop velocity of E7 versus applied voltage for different temperatures (45°C - 50°C – 55°C: nematic, 60°C: Transition, 65°C: Isotropic)

Afterwards, the influence of driving voltage applied to piezoelectric actuators on ejection of E7 droplets has been observed (Figure 3). Drop velocity increases linearly with applied voltage, with quite similar slopes, whatever the temperature is.

E7 reacts like a classical ink in the printhead when driving voltage is increased [7], which was not obvious at first, because of LC high sensitivity to an applied electrical field. Indeed, Negita [8] demonstrates that when an electric field of a few kV/ mm is applied, perpendicularly to the flow, on 5CB liquid crystal in its nematic phase, an increase in its apparent viscosity is observed. Then, the liquid starts to act like a non-Newtonian liquid. This electro-rheological effect is due to a change of the director orientation and determined by the balance between the electric field and the shear rate of the flow influences exerted on the molecules orientation. Owing to this electro-rheological effect, we may see a non-linear effect on drop ejection velocity of E7, as 5CB is its main component (51%).

Nevertheless, drop formation process is not disturbed here for several reasons. In XAAR318® printheads, driving voltage is not applied directly through the liquid, but across piezoelectric walls. As the electrodes are not isolated from the liquid, we can assume that local residual electric fields are seen by E7 in the channels. However, the electrical fields do not exceed 0,5kV/mm, which would exert only a small influence on E7 molecules orientation. Meanwhile, the piezoactuators create a high shear rate through the liquid that forces molecules to align with the flow whatever the electrical field is.

Moreover, the ejection frequencies used with these printheads vary between 1 and 4 kHz, whereas E7 response time to an electrical field application is about 35ms [5]. This does not give enough time to the LC molecules to align with the field and affect its apparent viscosity.

Thus, even if E7 might be sensitive to the electrical fields present around, its printing process is not affected. However, we can notice that at the transition temperature (60°C), the curve, shown figure 3, is lower than expected, which could be a consequence of the discontinuous jump of viscosity seen on figure 2.

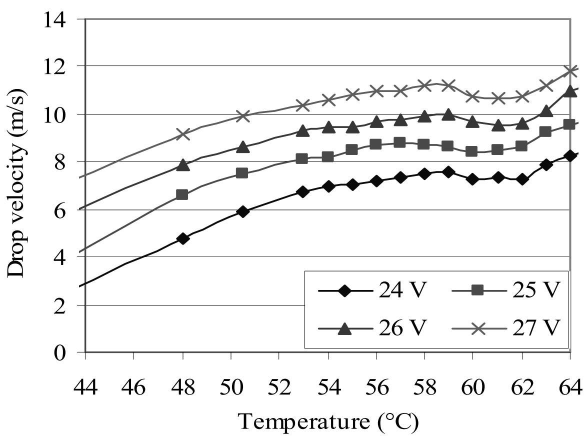

Figure 4 –Drop velocity of E7 versus temperature for different applied voltages

Then, we studied the influence of fluid temperature on drop velocity (Figure 4) for different driving voltages. As we could assume, drop velocity is rising gradually with the temperature, as the viscosity is falling [7]. Nevertheless, the curves present a discontinuous fall near the transition temperature. This proves that the sudden jump of viscosity is troubling the drop ejection.

Besides, above a critical value, the shear rate applied on a liquid crystal in its isotropic phase near TNI is known to create nematic areas in the isotropic phase [8]. This phenomenon is then changing the rheological properties of the E7 flow. Consequently, it should influence the phase transition that should happen at a higher temperature. In the inkjet process, the E7 is subjected to a very high shear rate (105-106 s-1) across the nozzle aperture, but, apart from the measurements accuracy, the results show that this shear rate does not shift the transition temperature of E7 (figure 4). As the nozzle hole has a really small volume comparing to the volume of the channel (>0,2%), we can assume that most of the energy transferred to the droplet ejected is determined in the channel, where shear rate is lower than in the exit hole. High shear rate is exerted for less than ten microseconds on the liquid through the hole, which should not be sufficient to create nematic areas in the E7 flow.

Far from T_{NI}, E7 is acting like a classical liquid, so the two rheological models (1) and (2) can be used to illustrate the variation of drop velocity when viscosity is increased (Figure 5). In both phases, the drop velocity decreases now linearly with the viscosity models, which is due to the augmentation of energy lost by viscous frictions during the ejection.

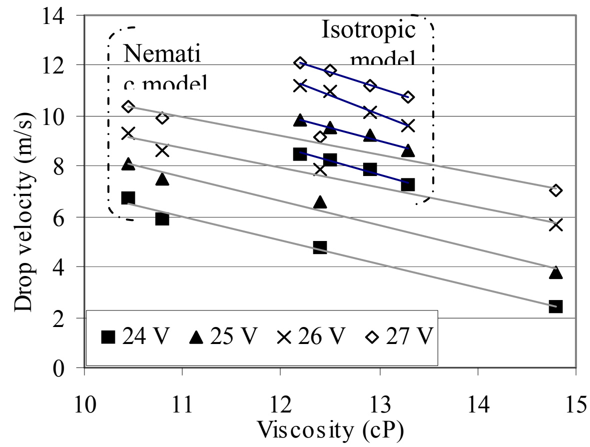

Figure 5 – Velocity versus viscosity of nematic model and isotropic model

Regarding these results, a simple experimental relation between ejection velocity (v) and printing parameters (driving voltage (U) and viscosity (η)) can be expressed, for each rheological model:

$$v = C_1 \times U + C_2 \times \eta + C_3 \quad (3)$$

With C1, C2 and C3 constant values depending on rheological properties of the liquid and printhead characteristics

Owing to the Arrhenius law connecting temperature and viscosity for Newtonian liquids, this expression can also be written:

$$v = C_1 \times U + C_2 \times \eta_0 \, e^{E/kT} + C_3 \quad (4)$$

With η_0, E two constant values depending on the liquid, k the Boltzman constant and T in Kelvin degree.

Then, experimental relations are expressed for both nematic and isotropic phases of E7:

-Nematic model:

$$v = 1{,}4\,U - 3{,}22.10^{-5} e^{4060/(\theta+273{,}15)} - 18{,}8 \quad (5)$$

-Isotropic model:

$$v = 1{,}18\,U - 8{,}3.10^{-3} e^{2530/(\theta+273{,}15)} - 5 \quad (6)$$

These expressions are useful to stabilize drop ejection process. Indeed, drop velocity can be fixed to a desired value (v_0), for instance to have no satellites, and equation (4) permits to calculate adequate driving voltage in function of E7 temperature in the printhead:

$$U = -\frac{C_2}{C_1}\eta_0 \; e^{E/k(\theta+273,15)} + \frac{v_0 - C_3}{C_1} \qquad (7)$$

This enables to have an optimized drop formation whatever the temperature is.

For E7, because of the viscous discontinuity, two relations are necessary to stabilize printing: relation (8) based on nematic rheological model (5) for temperature lower than TNI, and relation (9) based on isotropic rheological model (6) above:

-Nematic model:

$$U = 2{,}3.10^{-5} \; e^{4060/(\theta+273,15)} + 17{,}7 \qquad (8)$$

-Isotropic model:

$$U = 7.10^{-3} \; e^{2530/(\theta+273,15)} + 9{,}4 \qquad (9)$$

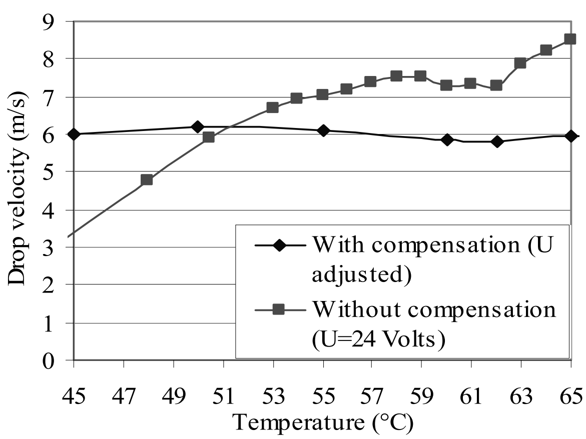

Figure 6 – Drop velocity versus temperature with (Aimed velocity 6m/s) and without voltage adjustments

Thanks to the application of equations (8) and (9) to adjust driving voltage, we managed to have a stabilized drop velocity around 6 m/s (± 5%), from 45°C to 65°C, while, without any adjustment, velocity of droplets varies between 3 and 8,5 m/s (Figure 6). However, we can notice that there are small variations around TNI, which shows that our models are not perfectly fitting the experience.

Moreover, the reading of live temperature in the liquid manifold of the printhead would enable us to adjust instantaneously the driving voltage, and compensate temperature variations that could occur in XAAR318® printhead, because of the heating of piezoactuators And PCB cards during high frequency prints [10].

Conclusion

After a presentation of particular temperature dependency of E7 viscosity in the nematic and the isotropic phases, the influences of inkjet printing parameters on drop formation have been detailed.

Thus, it has been demonstrated that electro-rheological effect disturbing the viscosity of thermotropic liquid crystals does not have any effect on drop ejection in XAAR318® printhead. However, the nematic-isotropic transition leading to a discontinuous jump in viscosity variation was presented to be potentially troubling for ejection of droplets. Then simple experimental models of both nematic and isotropic phases have been expressed to simplify the rheological behaviour of E7 liquid crystal. This enables us to adjust the driving voltage applied in function of the liquid temperature in the printhead, in order to stabilize drop velocity.

We can conclude that, in spite of complex rheological properties, the understanding of E7 behaviour in function of inkjet parameters influences enables us to have a stabilized and optimized drop formation of E7 with a piezoelectric DOD printhead. This makes E7 a perfect candidate to be accurately deposited in microelectronic systems with those printheads.

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