Electro-Rheological Model of HP Indigo ElectroInk

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Abstract. Electrolnk, developed and manufactured by HP Indigo Division for its digital printing presses, is a complex fluid of unique rheological and electrical properties. Depending on the shear and the electric field the ink may be solid- or fluidlike. As Electrolnk propagates in the press from the ink reservoir to the substrate, the concentration increases while the structure of the ink changes from a dispersion of non-interacting particles to an elastic solid. The authors' model treats Electrolnk as an interwoven structure of two continuous phases, one being a network of pigmented resin particles and the other the incorporated liquid. In an external electric field the network shrinks like a sponge and partial phase separation occurs. Application of the model is shown for the design of 100% transfer of ink from roller to roller in the press, which is necessary to meet the principal requirement of digital printing, that each printed page can be different. © 2013 Society for Imaging Science and Technology.

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INTRODUCTION

Hewlett–Packard Indigo Division develops and produces digital printing presses. These presses possess the capability that each page can be different from the previously printed page. This is made possible by Indigo's unique ink named ElectroInk. ElectroInk is a mixture of a dielectric fluid carrier, polymeric particles bearing colorants and additives. Due to some of the additives the polymeric particles acquire an electrical charge, allowing them to move under an external electrical field (Figure 1).

During the process the ink is transferred from roller to roller and its concentration increases from dilute dispersion in the ink reservoir to completely dry on the substrate (Figure 2). In order to ensure that each page can be different, 100% transfer of the colored part of the ink from roller to roller is required (i.e., no colorant can be left behind).

RHEOLOGICAL PROPERTIES OF ELECTROINK

Although the main purpose of the present article is to gain an understanding of how ElectroInk behaves in the printer, rather than advancing the understanding of its fluid behavior, to pursue our goal some knowledge of the most characteristic flow properties is necessary.

In dilute form, approximating the ink as non-interacting particles, which supposes only the presence of Stokes' law type forces produced by the carrier liquid, provides a reasonable description of the flow properties. At higher concentrations, however, starting at about 10% by volume, as in the transfer processes Xfer 0 and Xfer 1 (refer to Fig. 2), the particles are strongly interacting and the rheological model of the ink should be quite different. Experiment shows that at these concentrations the ink exhibits finite yield stress to flow. Figures 3-5 show the results of oscillatory flow tests carried out in a TAI AR-2000 rheometer using parallel plate geometry. In such experiments the shear stress applied to the fluid varies with time according to a sinusoidal law and the shear rate is measured. The ratio of the stress and the shear rate is expressed through the complex modulus (or alternatively complex viscosity).¹ The real part of the modulus referred to as the storage modulus is related to the elastic contribution of the deformation whereas the imaginary part referred to as the loss modulus is related to the plastic part. The ratio of the two, referred to as $tan(\delta)$ or loss tangent, characterizes the state of the given material: δ is zero for fully elastic solids, 90° for ideal fluids and between zero and 90° for visco-elastic substances.

At small shear the ink, like solids, exhibits high modulus and low loss tangent (low δ) in oscillatory experiments. When the shear is larger than a certain value referred to as the yield stress, the modulus drops, the ink starts flowing and δ goes up. The process is reversible: running the same test on the same sample a second time the transition from solid- to fluidlike behavior appears again (Fig. 4).

The yield stress increases sharply with increasing concentration (Fig. 5).

Sedimentation experiments show that the model of non-interacting particles is not correct. If there were no interaction between the particles then the final concentration of the sediment would be equal to some most dense packing independent of the starting concentration and the acting force field. In contrast to this, experiments have shown that the concentration of the sediment increases logarithmically with both the speed of centrifugation and the initial concentration.

THE TWO-CONTINUOUS-PHASES ("SPONGE") MODEL

In order to explain such behavior we developed a theory named the two-continuous-phases model. The model describes the structure of ElectroInk in terms of two interwoven continuous phases, one being an elasto-viscous solid-like substance made of strongly interacting particles and the second phase being the pure solvent.

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Figure 1. Low concentration ElectroInk-schematic.



Figure 2. Ink transport in the press-schematic.



Figure 3. Oscillatory flow test of medium concentration (actually 16.8 %v) of cyan Electrolnk (square markers) at 10 Hz. Solid lines represent the storage modulus while dashed lines relate to δ (delta). For comparison the behavior of an ideal fluid (mineral oil used for calibration-MO-circle markers) and an almost perfect elastic material (polyurethane rubber-PU-triangle markers) is also shown.



Figure 4. Repeated flow test of the same ink as in Fig. 3. The filled markers denote the second run. Solid lines represent the storage modulus, dashed lines relate to δ (delta).

The transition from the approximation of a dilute dispersion of non-interacting particles to the two-continuousphases model can be schematically illustrated as shown in Figure 6(a)-(c). We start with diluted ink of randomly dispersed non-interacting particles (Fig. 6(a)). As the concentration increases (Fig. 6(b)) the particles get closer to each other and the increasing interaction between them creates a structure (pictured schematically in Fig. 6(c) as a network).

The influence of an external force field on the ink depends on the direction of the force. Shear stress and tension may destroy the network of the particles and lead to solid-to-fluid transition. Experiment has shown that such change is reversible (refer to Fig. 4), i.e., the network reforms again. Compression forces do not destroy the network. The influence of a compression force field on the ink is treated as follows. In real cases some external mechanical barriers (e.g., the non-penetrable surfaces of the rollers) are imposed on opposite sides of the mixture that prevent it from moving as a whole (Figure 7(a)). By applying an external volume



Figure 5. Variation of the yield stress with concentration for cyan ink. Each point on the graph is derived from independent measurements described in Fig. 3.

field to the particles the "network" is pushed to one of the barriers and gets deformed (Fig. 7(b)). The external field can be both gravitational and electrical. Gravitational field exerts pressure on the network when the particles have higher density than the surrounding medium (which is usually the case). Electrical field acts directly on the particles which bear net electrical charges. In computations the network is replaced by an elasto-viscous continuum (Fig. 7(c)).

Thus, in contrast to dilute ink modeled as a liquid in which solid particles are dispersed, the two-continuousphases model treats the ink rather as a solid in which the liquid is dispersed. The elasto-viscous "solid" part behaves like a wet sponge: when pressed it releases liquid (hence the nickname "sponge model"). To calculate any deformation the techniques of continuum mechanics and electrodynamics should be applied. For example, sedimentation can be modeled as the deformation of the "sponge" under its own weight, i.e., the upper layers exert pressure on the bottom layers (Figure 8). As a given layer gets compressed its concentration increases, involving an increase of the yield stress. When the yield stress reaches the value of the local hydrostatic pressure the compression stops. Thus the concentration of the sediment through the hydrostatic pressure is a function of the starting concentration and the speed of centrifugation and increases in the direction of the gravitational field.

ELECTRO-RHEOLOGICAL EXPERIMENTS

Electro-rheological measurements were carried out in a TAI AR-2000 rheometer using parallel plate geometry and applying voltage to the plates (Figure 9). Typical results obtained for medium concentration ink are shown in Figure 10.

As one can see, the apparent viscosity decreases when voltage is applied across the ink. Actually, what happens is that the solid part ("the sponge") bearing static electrical charges gets squeezed under the influence of the applied field (like under its own weight in sedimentation experiments) and releases some of the absorbed liquid, as schematically shown in Figure 11. Since the liquid has a much smaller



Figure 6. (a)–(c) Building the model (explanation in the text).



Figure 7. (a)–(c) Building the model–application of an external force field.



Figure 8. Sedimentation: layers are compressed under the weight of the layers above.

viscosity the total viscosity decreases. As the voltage is increased the change may be smaller since the "sponge" is now denser and accordingly harder.

A simple and useful approximation is to assign a certain viscosity to each phase. In this way the relative thicknesses of the phases can be obtained. Since the stress is constant across the sample the strains will add and

$$1/\eta_{\text{apparent}} = (1 - X)/\eta_{\text{concentrate}} + X/\eta_{\text{liquid}}, \qquad (1)$$

where $\eta_{apparent}$, $\eta_{concentrate}$ and η_{liquid} are the measured viscosity, the viscosity of the concentrate and the viscosity of the pure liquid respectively, and *X* denotes the relative thickness of the liquid phase.

It is useful to introduce the response coefficient of the ink to external voltage, defining it as the ratio of the relative



Figure 9. Electro-rheological measurement: voltage is applied to the plates.



Figure 10. Change of apparent viscosity with electrical bias. Viscosity was computed from the torque measured on cyan ElectroInk of 14.5 %v at a constant shear rate of 100 s^{-1} .

thickness to the electrical field strength:

$$k = X/E. \tag{2}$$

Taking $\eta_{\text{concentrate}} = \eta_{\text{apparent}}$ at zero voltage and $\eta_{\text{liquid}} = 1.25 \text{ cP}$, the data presented graphically in Fig. 9 yield a value of k = 1.5e - 8 m/V.



Figure 11. Interpretation of the electro-rheological measurement-schematic.



Figure 12. Electrical transfer—schematic: (a) entering the nip; (b) in the nip; (c) exiting from the nip.

APPLICATION OF THE TWO-CONTINUOUS-PHASES MODEL TO THE ELECTRICAL INK TRANSFER PROCESS

The transfers of ink in Xfer 0 and Xfer 1 (refer to Fig. 2) are electrically induced processes. In Xfer 1, for example, the ink arrives at the photoconductor drum and goes over to the intermediate transfer drum. The transfer process can be broken down into three steps (Figure 12).

The ink arrives at the contact of the drums as a homogeneous material. A voltage is applied to the intermediate drum relative to the photoconductor. Thus, between the photoconductor drum and the intermediate transfer drum the solid part of the ink containing the colorants gets squeezed and releases some liquid. Downstream of the contact area the drum surfaces separate and the low viscosity liquid splits into two layers. One layer goes with the photoconductor drum but, with adequately chosen parameters, this liquid does not contain any colorants (see the simulations below).

A simple simulation of splitting at the exit supposes only two layers, one containing the solids and the second being the pure liquid. Since in splitting shear stresses are present,² the layer of solids can also be treated for simplicity's sake as a fluid but of much higher viscosity than the pure liquid. In order to satisfy the requirement for conservation of mass while having non-slip conditions on the surfaces, as the separation between the drum surfaces increases the velocity of the fluid has to vary across the gap. Splitting will occur at the stagnation point where the velocity is equal to zero. On the interface separating the layers the shear stress is continuous and the velocities of the two layers are equal.

Simulations were carried out in the "lubrication" approximation.² Some typical results based on a viscosity ratio of 400 between the phases can be seen in Figure 13, where the calculated velocity profiles in the gap are shown during exit from the contact area. The plots have the following meaning. *Y*-axis: the relative distance between the drums, which is 0 at the intermediate transfer drum, 1 at the photoconductor drum; *X*-axis: the relative velocity across the gap (1 at the drums and zero at the locus of the splitting).

When the liquid phase is thick enough the splitting occurs in the liquid. Since the liquid is free of colorants the transfer to the intermediate transfer drum is 100% and stable (Fig. 13(a)). As the relative thickness of the liquid decreases the splitting point moves to the interface (Fig. 13(b)). This corresponds to 100% but unstable transfer. When the relative amount of the liquid is less than a certain critical value (in this case 5%) the splitting occurs in the solid phase and less than 100% of the colorants are transferred to the intermediate transfer drum (Fig. 13(c)).

DISCUSSION

Although the model is proposed for use at medium concentrations (approximately 10%–30% by volume), theoretically it is valid for all concentrations. At other concentrations another model may be more convenient for performing



Figure 13. Velocity profile in the gap. Layer (1): liquid (dashed lines). Layer (2): solid phase containing the pigments (solid lines). Viscosity ratio: 400. (a) Liquid phase: 10%, splitting in the liquid phase—stable 100% transfer; (b) liquid phase: 5%, splitting on the interface—unstable 100% transfer; (c) liquid phase: 1%, splitting in the solid phase—less than 100% transfer.

computations, e.g., the "Stokes model" may be more convenient for treating very dilute dispersions.

We have to make here a comparison between phenomenological (macroscopic) and structural (microscopic) theories. The model proposed here is a phenomenological one. Other authors have published work on the structural approach of ink or toner consolidation.^{3–5} Structural models suppose some well defined structure and interaction forces between large numbers of individual particles.

The relationship between the phenomenological model and structural approaches is like the relationship between thermodynamics and statistical physics. The phenomenological model, like thermodynamics, does not enter into the details of the structure and the interactions between the structural units. The structural approach needs the knowledge of a large number of parameters, notably particle size and shape and their distribution, electrical charge and its distribution, mechanical properties ("hardness") of individual particles, the interaction forces between the particles (which can originate from as many as five sources⁶), and the influence of temperature, pigments and additives. In most cases these parameters cannot be measured directly; more or less justified assumptions are made about their value (e.g., taking mono-disperse spherical particles). ElectroInk particles are neither spherical nor mono-disperse. Thus, results obtained this way always bear some ambiguity.

In contrast to this, phenomenological models introduce only a small number of experimentally measurable material characteristics. In our model the "network" between the particles is only used as a descriptive tool to visualize the presence of an interaction between the particles. The crucial step in building our model is the replacement of the "network" by a continuous medium (Fig. 7(b) and (c)), thus smearing out the exact form of the interactions. This renders the model phenomenological. The choice of model depends also on the purpose of the application. The virtue of the phenomenological model is that while being extremely simple it is capable of interpreting and connecting a wide range of experimentally observable phenomena, e.g., rheological and electro-rheological properties, sedimentation, electrical consolidation and processes occurring in the press, e.g., ink transfer.

Structural models are useful when they give the basis for a macroscopic or phenomenological description. It is only through structural theories that the influence of the particle properties, temperature, surface active agents and external force fields on the ink's macroscopic properties can be assessed. In terms of rheology a microscopic theory taking into account the exact form of the interactions should provide the constitutive equations of the solid phase. This is a subject of further study.

It has to be noted that conventional electro-rheological fluids behave differently; in contrast to ElectroInk they exhibit viscosity increase in an electrical field.⁷ The reason is that most electro-sensitive fluids contain particles that possess electrical dipoles rather than net charge. Thus, in an external electric field they only rotate and orient themselves along the field. This leads to building "bridges" between the plates of the viscometer.⁷ In contrast to this, in ElectroInk the particles possess non-zero electrical charge which results in much larger forces than the dipole–electrical field interaction and in linear displacement of the particles rather than rotation.

It has to be emphasized that the above simulation of the ink transfer process from the photoconductor drum to the intermediate drum does not pretend to be new and was presented here only for the purpose of demonstrating how the "sponge" model can be applied to understanding processes on the press. Ink splitting is an extremely important process in printing and is well investigated.⁸ The application of fountain solution creates a situation very similar to the model computed here.⁹ Our simulation can only be considered as a first approximation and not an exact solution. Important properties such as surface tension, cohesive strength, elastic deformation and extensional viscosity were omitted in this analysis. Despite this, it proved to be rather successful in interpreting phenomena occurring in Indigo presses. For example, experiment shows, in full accordance with the model, that a certain minimum electrical bias is needed for perfect transfer from the photoconductor drum to the intermediate drum (Figure 14).

Although the experimental data presented in this article were mostly collected for cyan ink they are representative of ElectroInk. Other colors and ink formulations gave similar results.

CONCLUSIONS

ElectroInk, consisting of pigmented polymer particles dispersed in carrier liquid and several additives, is a complex fluid with unique rheological and electrical properties. Its mechanical behavior depends on concentration, temperature, and the magnitude and rate of change (or



Figure 14. Efficiency of the transfer from the photoconductor drum to the intermediate drum as a function of the applied electrical bias for yellow ink. Markers denote experimental values.

frequency—in the frequency domain) of the applied stress. In an external electric field the polymer particles acquire electrical charge so that an electrical field also influences the rheological behavior. Depending on the shear and the electric field the ink may be solid- or fluidlike. As ElectroInk propagates in the press from the container to the substrate, the concentration increases while the structure of the ink changes from a dispersion of non-interacting particles to an elastic solid. The theory presented here was developed for medium concentration ElectroInk and models it as a structure of two interwoven continuous phases, one being a solid-like network of pigmented resin particles and the other the incorporated liquid. In an external electric field the network shrinks like a sponge and partial phase separation occurs. Although the model is phenomenological, without going into the details of the interactions between the ink particles, it is capable of explaining such phenomena as concentration variation in sedimentation and variation of viscosity under electrical field. It yields practically important parameters, such as the response coefficient of the ink to external voltage, which gives the relative thickness of released liquid by electrical field.

Application of the model is shown for the design of 100% transfer of ink from roller to roller in the press, which is necessary to meet the principal requirement of digital printing, that each printed page can be different.

Development of a microscopic theory that takes into account the exact form of the interactions between the ink particles and yields a quantitative constitutive equation of the solid part in particular, and ElectroInk in general, is the subject of further study.

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REFERENCES

- ¹T. Mezger, *The Rheology Handbook* (Vincentz Verlag, Hannover, 2002), ISBN: 3-87870-745-2.
- ² S. Middleman, Fundamentals of Polymer Processing (McGraw-Hill, Inc., 1977), ISBN: 0-07-041851-9, Chapter 7.
- ³ H. Mizes, "The structure of toner sediments simulated with random ballistic deposition," *Proc. IS&T's NIP15: Int'l. Conf. on Digital Printing Technol.* (IS&T, Springfield, VA, 1999), pp. 495–498.
- ⁴ S. Chang, P. Rameshand, J. LeStrange, J. Domoto, and J. Knapp, "Stabilization of liquid developed images," *Proc. IS&T's NIP15: Int'l. Conf. on Digital Printing Technol.* (IS&T, Springfield, VA, 1999), pp. 638–641.
- ⁵ S. Chang, H. Till, E. Viturro, K. Watson, A. Pérez, A. González, and A. Castellanos, "Measurement of toner cohesion in liquid inks using a visualization cell," *Proc. IS&T's NIP14: Int'l. Conf. on Digital Printing Technol.* (IS&T, Springfield, VA, 1998), pp. 218–221.
- ⁶ L.-H. Lee, Adhesive Bonding (Plenum Press, New York, 1991), p. 22, ISBN: 0-306-43471-7.
- ⁷ R. G. Larson, The Structure and Rheology of Complex Fluids (Oxford University Press, New York, 1999), ISBN: 0-19-512197-X), Chapter 8.
- ⁸ See for example: H. Kipphan, Handbook of Print Media: Technologies and Production Methods (Springer, 2001), ISBN: 3540673261.
- ⁹ See for example: A. Glassman, *Printing Fundamentals* (TAPPI Press, 1985), p. 326, ISBN: 0-89852-045-2.