

PAPER

# Machine Learning for Ink Optimization for Printed Electronics: A Perspective

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## ABSTRACT

Printed electronics, embracing the ability to achieve electronic technologies on surfaces and geometries of varying complexities, have captured massive attention over the past decades. At the heart of the printed electronics is the ink that is used. This perspective article sheds light on the use of machine learning (ML) in optimizing the formulation and the processability (e.g., deposition rate, sintering, etc.) of such inks with the aim of achieving printed traces and components with desired conductivity and structure (e.g., structures with reduced voids, cracks, and coffee stain effects).

## KEYWORDS

machine learning (ML), printed electronics, conductive inks, metal nanoparticles, voids, coffee stain effect

## 1 INTRODUCTION

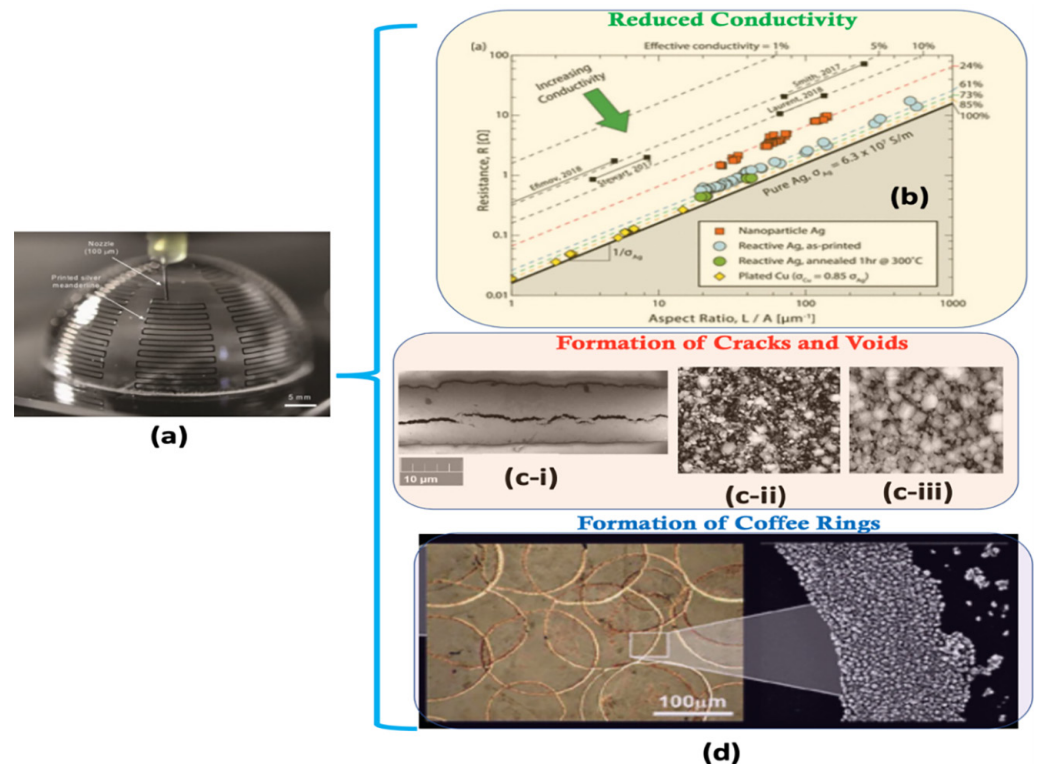
Technologies reliant on conventional electronics impact every sphere of our daily life and livelihood. While conventional electronics work best for rigid and flat surfaces, they might be unsuitable for more niche applications (e.g., flexible and wearable sensors). This is where “Printed Electronics” become relevant. Printed electronics refer to electronic systems that are “printed” or “deposited” [1]. Under typical conditions, such electronics are fabricated by depositing a conducting ink that, when dried (or sintered), will lead to the formation of traces and lines and architectures that are electrically conducting. Given that one cannot “print” molten metals (due to their very high temperatures), the main way forward is to work with an ink that contains conductive metal nanoparticles (e.g., silver or copper nanoparticles or NPs): this ink gets deposited, the liquid evaporates (dries), and one is left with traces of conducting printed silver or copper (or aggregated silver or copper NPs) [2]. While this ability to “deposit” and achieve conductive traces (this is what “Printed Electronics” is built upon) enables achieving electronics for complicated architecture (e.g., electronics for wearable glucose sensors) [3], the very fact that

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it is the aggregated NPs that trigger the conductivity causes several limitations. Some of these limitations are smaller conductivity (as compared to that achieved by conventional electronics working with liquid metals) [4], inherent porosity [5], voids [5], and cracks [6] in the printed traces (and the associated reliability failures), formation of the coffee ring effect [7], etc. (see Figure 1 for a summary). Therefore, it is intuitive that there is a massive scope for designing and engineering the ink needed for printed electronics applications with the aim of optimizing various objectives, such as improving the conductivity, reducing the voids and porosity of the traces, reducing the coffee stain effect from the dried traces, etc. Given such a multi-objective optimization challenge, machine learning (ML) and different regression methods (developed using some of the existing experimental results as training data) will be extremely useful. While there has been considerable research in developing ML methods for optimizing the printing strategies for printed electronics applications [10] or for developing other types of inks [11], there has been a lack of papers on optimizing the formulation of highly conductive inks with exceptional features (e.g., small sintering temperature, absence of coffee ring effect, significantly reduced crack and void formation). There have been only a handful of papers that have aimed to optimize the ink for printed electronics using the ML (discussed later), and almost none of them leveraged the appropriate understanding of the involved physics in their optimization efforts. Thus, there is a significant gap and hence a tremendous opportunity in using ML methods for optimizing inks that enable desired outcomes in printed electronics applications: this perspective article sheds light on that.



**Fig. 1.** Pictorial summary of key challenges of the ink for printed electronics

*Notes:* (a) Example of printed electronics: conducting traces being printed on a hemisphere [8], (b) Examples of reduced conductivity (or enhanced resistance) [11], (c-i) [15], (c-ii,iii) [14], (d) [9].

## 2 CHALLENGES WITH INKS FOR PRINTED ELECTRONICS: HOW MACHINE LEARNING CAN HELP?

Printed electronics aim to fabricate (via 3D printing) devices that can replicate the functionality of conventional electronics on surfaces with complicated topologies. For example, one can print resistors, interconnects, sensors, antennae, capacitors, etc. on highly curved surfaces. In fact, such capabilities enable integrating electronics to different organs, biosystems, chemical systems, and mechanical systems much more seamlessly. While the usefulness of printed electronics is not to be doubted, unfortunately most of our electronic products (from cell phones to laptops) still heavily rely on conventional electronics. There are several factors responsible for such a scenario, with all of them being related to the ink that is being printed. Below we highlight some of these issues.

### 2.1 Reduced conductivity associated with printed electronics

Given that the printed electronics are based on the aggregate of conductive NPs, the conductivity of this NP aggregate is the overall conductivity of the printed electronics system and hence much smaller than the conductivity associated with molten metals. For example, it is commonplace to have a conductivity of a trace printed with silver NP-laden ink to be only 10–20% of the bulk metallic silver [4, 12]. In fact, less careful sintering (e.g., sintering at a reduced temperature or sintering too fast) might lead to even smaller conductivity values.

### 2.2 What causes this reduction in conductivity?

It has been widely believed that the intrinsic voids that result from the sintering-driven formation of aggregates of conductive metal NPs (as described above) reduce the conductivity. For example, Rosker et al. [4] demonstrated that the conductivity of the sintered traces ( $\sigma_{sin}$ ) can be related to the void fraction ( $\phi_{void}$ ) inside the sintered traces as:

$$\sigma_{sin} = \sigma_m \left[ \frac{2(1 - \phi_{void})}{2 + \phi_{void}} \right], \quad (1)$$

where  $\sigma_m$  is the conductivity of the bulk metal.

In a recent study [12], we conducted sintering experiments on printed silver NP-laden conductive traces (printed with commercial silver ink) and found that the  $\phi_{void}$  can be as small as 12%, which will lead to  $\sigma_{sin}$  to be almost 83% of  $\sigma_m$ ; in other words, the conductivity of the sintered traces is very close to the conductivity of the bulk metals. In our paper, we demonstrated that the main cause of conductivity reduction is not the presence of voids, but the residual polymer layer (remnants from the ligand layers with which the NPs in the ink are grafted for stability) on the surfaces of the NPs or nanoaggregates (formed from the NPs) that remains present even after sintering. Of course, the greater the sintering temperature, the more prominent is the removal of this residual polymeric layer, and therefore, the greater the improvement in conductivity. However, very often this sintering temperature cannot exceed values of 150°C or 200°C owing to the nature of the underlying substrates; as a result, the coupled effect of the presence of voids and the presence of the residual polymeric layers continues to hinder the conductivity of the printed electronics.

### 2.3 Coffee stain effect

One of the most fundamental colloidal dynamics events associated with the evaporation of a colloid-laden drop is the “coffee stain” effect [13]. When a coffee drop evaporates, the coffee particles follow the streamlines of the evaporation-driven liquid flows and get deposited at the three-phase contact line, forming a ring: this effect is known as the “coffee ring” or “coffee stain” effect. Thus, considering the metal NP-based ink as a colloidal solution, it is quite intuitive that such a coffee stain effect is invariably encountered during the sintering of the deposited (printed) NP-laden ink [7]. Such a coffee stain effect is also known to trigger deformities in printed components. In a seminal paper, Yunker et al. showed that while spherical particles invariably led to the formation of the “coffee stain” effect, ellipsoidal particles can actually arrest coffee stain effect [14]. This idea has been at the core of developing commercial metal NP-based inks with flaky (non-spherical) shapes. Flakes are extreme forms of ellipsoids, and therefore, it is expected that the coffee ring effect will not occur in traces printed with such nano-flaky inks. In fact, we have ourselves leveraged this principle of size-based arrest of the coffee stain effect in developing a novel metal-free conductive ink that can be used for printing temperature and humidity sensors [7]. The ink consists of different percentages of CNT (carbon nanotube) and GO (graphene oxide). The CNTs ensure the ink is conductive, while the presence of the flaky GO ensures that printed CNT-GO ink does not demonstrate the coffee stain effect.

### 2.4 Reliability issues with the printed traces due to the presence of voids and aggregates

Printed electronics are characterized by printed conductive traces composed of metal NP (silver NP) aggregates. The formation of such aggregation invariably introduces significant voids and cracks [5, 6], or locations of fundamental weaknesses across the printed traces. Accordingly, when these traces are subjected to different environmental traces, these spots of weakness give in, and there occurs permanent damage to these printed traces.

### 2.5 Examples of how machine learning can help?

The limitations associated with the inks used for the printed electronics, as have been discussed above, are intricately connected to the ink composition (e.g., NP shape and structure, the nature of the ligands stabilizing the NPs, etc.) and the process dynamics (e.g., the sintering temperature or profile, etc.). Therefore, there is a scope of optimization of these properties and processes, and that is where ML comes into play.

For example, consider the case where one needs to optimize three things: (1) particle loading inside the ink, (2) the nature and grafting density of the ligands on the particle (NP) surfaces, and (3) the sintering profile (that does not exceed 150°C). The target is optimizing the conductivity of the printed traces. Thus, this is an example of a single objective optimization. One can apply various strategies of supervised ML methods for the optimization and identification of the combinations of these three parameters (see above) that ensure the best conductivity. These ML methods can be Bayesian optimization (for situations where the amount of initial training data is relatively less) neural networks, or other regression methods.

Let us consider another situation where one would like to simultaneously target optimizing (1) conductivity of the printed traces and (2) the arresting of the coffee stain effect. For such a scenario, the input conditions are as follows: (1) the concentration of the NPs inside the ink; (2) the shape of the NPs (purely spherical, purely flaky, and a combination of spherical and flaky with different weight percentages); (3) the nature and grafting densities of the stabilizing ligand; and (4) the sintering profile. Here the problem is a multi-objective optimization (MOO). There have been previous studies performing such MOO in the context of developing printable inks using ML methods such as Pareto plots [11]. One can apply similar strategies here as well and ensure optimizing the different input conditions for simultaneously optimizing the two stated objectives.

### 3 EXAMPLES OF STUDIES USING MACHINE LEARNING FOR INKS FOR PRINTED ELECTRONICS

The Table 1 below provides a list of papers (and their main contributions) that have employed ML for problems involving conducting inks for printed electronics applications. These papers, however, do not specifically focus on optimizing the formulations of such inks.

**Table 1.** ML for problems involving conducting inks for printed electronics applications

Reference	Problem Studied and Main Output	ML Methods Used	Ink Used
[15]	Use of ML for optimizing input conditions for ink jetting in inkjet printing of electronics. Main Output: Identifying the drop velocity and drop radius associated with ink jetting for different inks	Various regression models, including decision trees (DTs) (boosted DTs and random forests)	Silver nanoparticle ink (ANP DGP 40LT-15C) (Advanced Nano Products, Co., Sejong, Korea)
[16]	Use of ML for automated ink selection for printed electronics. Main Output: Preferential automated selection (avoiding manual trial-and-error approach) between three types of existing conductive inks	Multilayer Perceptron Neural Network (MLPNN) and Particle Swarm Optimization (PSO), named PSO-MLPNN	Conductive Inks (Carbon, Copper, Silver NPs-Laden Inks)
[17]	Use of ML to ensure that printed RFID antennas can be used for liquid sensing. Main Output: Sensing the content of colorless and unidentified liquids	Two-layer back propagation pattern recognition-based neural networks	Conductive graphite nanoplates (GNPs) ink
[18]	Use of an ML-based architecture for microwave characterization of inkjet-printed components on flexible substrates. Main Output: Quantification of the ink conductivity and dielectric properties of the printed components	Regression models (Decision Tree (DT), eXtreme Gradient Boosted Trees Regressor (XGB), Light Gradient Boosting (LGB), ResNet, SVM, CNN, RNN)	Silver nanoparticle ink

Interestingly, in addition to the research investigating the use of ML for the formulation of ink for printed electronics, there is one study that has focused on utilizing machine learning to select conductive inks for various printed electronics applications [19]. An automatic ink selection system rather than relying on manual intervention for printing applications is proposed in this study, where initially a

novel artificial neural network (ANN) framework is designed. Further on, multilayer perceptron neural network (MLPNN) is created to select the conductive ink based on input characteristics such as product life, quality, usage, and handling, grams per square meter (GSM), caliper, brightness, tear resistance, and moisture content. Three types of conductive inks are considered, such as carbon, copper, and silver inks. The proposed study successfully demonstrated the effectiveness of the MLPNN to automate and improve the decision-making process in the selection of conductive inks for printing applications.

## 4 SCOPE OF FUTURE WORK

The significant lack of papers on the use of ML in improving/optimizing the properties of the ink that is used for printed electronics can be attributed to (1) over-reliance on the commercially available ink for printed electronics and (2) lack of understanding of a lot of intricate physics and chemistry that dictate the behavior of such inks.

Our extensive previous work on identifying different aspects of conducting inks [5–7] as well as significant work on the use of ML [20–23] for various engineering and soft matter systems place us well to identify the future problems where ML can significantly improve the formulation of the ink for printed electronics. Below we provide some specific examples. It is worth mentioning here that the specific examples of various ML methods (e.g., ANNs or Bayesian optimization or support vector regression (SVR)) that will be suggested below for improving the different aspects of the ink formulation are those that have not been so far applied to experiments for optimizing the properties of conductive inks. Rather, these methods have shown extremely promising applications with regard to experiments for other systems. We believe, as elaborated below, that these methods will be useful if they are properly employed for optimizing the properties of the conductive inks.

### 4.1 Formulating ink that shows high conductivity at a reduced temperature

Typically, after printing, sintering is conducted to ensure (1) the liquid evaporates and ensures self-assembly of the NPs and (2) the ligands grafting the NPs get decomposed (and removed) so that the NPs can come in direct contact with one another, thereby increasing the conductivity. However, our paper showed that at typical sintering temperatures ( $\sim 150^\circ\text{C}$ ), some amount of this ligand remains unrecovered, which in turn reduces the conductivity [12]. Of course, if the sintering is carried out at higher temperatures, the ligand removal is much more complete, thereby increasing the conductivity. However, the problem with such an endeavor is that the sintering temperatures become so high that they are unusable for typical substrates where the printing is conducted. Under these circumstances, an ideal option will be formulating the choice of ligands in a manner such that (1) they adsorb easily on the NPs and (2) detach quickly from the NPs at relatively lower temperatures. Methods such as descriptor-based artificial neural networks (ANN) have been previously used to demonstrate a new property of existing systems based on training data derived from the experimental results on those systems. For example, ANN models have been built on training data derived from the experimental results on anti-fouling properties of polymer brushes to yield novel anti-fouling properties of existing brushes [24]. Given that the ligands on the NPs behave as polymer brushes, we can

use such descriptor-based ANN models (previously used for optimizing applications of polymer brush-based systems [24]) to study novel detachment behavior of the ligands on the NPs (of the conductive ink) as a function of the formulation of the ligands, with the ANN model being developed using the training data set consisting of data on the conductivities as a function of temperature of different inks containing NPs with different types of ligands. On the other hand, in case we are interested in developing new ligands for the NPs (or ligands that will detach at a much lower temperature), we can use methods such as SVR model. Previous studies have shown the use of such SVR methods in designing new brushes with antifouling properties [24]: here, one can use such SVR methods to design new ligands with desired detachment behavior (i.e., detachment at significantly smaller temperatures).

#### **4.2 Ink that causes traces with the lowest voids and cracks after printing**

Another aspect of concern is the inevitable voids that are formed in the sintered traces printed with conducting inks [5]. These voids lead to reduced conductivity (as discussed above) and also cause reliability failures of the printed traces (when subjected to different environmental stresses). Another issue is the formation of lateral cracks in traces printed with conducting inks. Both these issues are related to the formulation of the ink and the manner in which (a) NPs are deposited (e.g., the thickness of each deposition layer and consequently the rate of printing) and (2) NPs respond to the sintering (or conversely, the rate of sintering). The challenge is that while the first issue can still be regulated, there is little control over the second issue. Hence here too, ML can help. One can consider a particular ink and study how the different deposition rates and the sintering profiles affect the formation of such voids and cracks. It's a multi-objective optimization problem (optimizing the rate of deposition and rate of sintering), and therefore, some of the most standard methods of multi-objective optimization [e.g., Bayesian optimization (BO)] can be useful. For example, following Chen et al. [25], one can employ BO to simultaneously optimize the rate of deposition (printed layer thickness) during Direct Ink Writing as well as the sintering profile that ensures desired distributions of voids and aggregates. Like other supervised ML methods, here too one would first need to create the appropriate training data set (a data set that for a given conductive ink outlines the distribution of cracks and voids as functions of the deposition rate and sintering profile) on which the BO will train and accordingly predict the appropriate combinations of deposition rate and sintering profile for printed traces with insignificant cracks and voids. Interestingly, for a given ink, this particular problem is equivalent to the problem of multi-objective process optimization for 3D printing processes, and therefore, other ML methods [e.g., methods such as K-means clustering, principal component analysis (PCA), reinforced learning, ANN, k-nearest neighbors (KNN), convolutional neural network (CNN), etc.] that are routinely employed for optimizing 3D printing processes can be utilized here as well.

#### **4.3 Ink that causes traces with the least coffee stain effect**

The presence of the spherical NPs in the ink leads to the coffee stain effect when the ink is printed. Such a coffee stain effect leads to unevenness in the thickness of the printed traces, issues with the reliability of the traces, etc. There have been some efforts in formulating inks with nanoflakes (nano ellipsoidal silver NPs) in order to

ensure the arrest of the coffee stain effect [26]. However, there is little knowledge on how the conductivity as well as other process abilities (e.g., sintering response) of the traces with such nano flaky ink varies as compared to the conductivity of the standard ink with spherical NPs. Under these circumstances, one can conduct experiments with ink containing a mixture of spherical and nano flaky (nano ellipsoidal) NPs and see how the conductivity, the coffee ring formation, and voids and cracks on the printed traces vary. This problem again is a problem of multi-objective optimization (trying to optimize several objectives simultaneously). One approach can be a standard multi-objective optimization (MOO) procedure based on the construction of the Pareto sets (see Erps et al. [11]). In this paper [11], the authors played with the components of polymeric inks in order to formulate an ink that simultaneously optimized various mechanical properties of the components printed with such ink. For the present case, the same problem of optimizing the components of the nano inks (namely the relative percentage of the spherical and ellipsoidal nanoparticles) is in question; hence, we shall use the ML-based MOO procedure (e.g., use of Pareto sets) for fabricating ink with an appropriate relative composition of flaky and spherical ink in a manner such that the multi-objectives (prints with high conductivity, prints with minimum coffee stain effect, and prints with the least voids and cracks) can be optimized.

#### 4.4 Cases with other types of conductive inks

As can be noted, so far in the paper we have mostly focused on standard commercially available conductive inks. Of course, there has been extensive research in formulating conductive inks in the laboratory setup. For example, we developed CNT-GO (CNT: Carbon Nanotubes; GO: Graphene Oxide) inks that were conductive (due to CNT), led to a uniform deposition (due to GO), and were used to print temperature and humidity sensors [7, 27, 28]. Of course, one can perform different functionalizations of GO to make it more conductive without losing the printability and stability of the CNT-GO (or more specifically CNT-*f*GO, where *f*: functionalized): several experiments followed by ML-driven optimizations can be conducted for this purpose.

Similar things are possible for other types of in-lab-formulated non-commercialized conductive inks that are based on some combination of conductive metallic particles (e.g., silver) and non-metallic conductive particles (e.g., CNT). Several review articles [29, 30] provide detailed information on such inks and the associated nanomaterials.

Given that the overall research community does not have seamless access to these specific custom-built (or in-lab fabricated) inks, it is difficult to perform extensive experimentation with these inks to develop the necessary training data set for employing ML-based optimization. Accordingly, we focus on discussing mostly commercial inks and the manner in which ML can be employed to improve their performances.

## 5 CONCLUSIONS

In this perspective article, we shed light on the challenges associated with the conductive inks for printed electronics applications and the manner in which different ML methods can help address these challenges. We first introduce the main

challenges associated with such inks, often stemming from the very fundamental nature of the metallic or conductive NPs that constitute the inks. Subsequently, we discuss some previous papers, which are relatively few in number, that have employed ML methods in optimizing such ink formulation and process abilities. Finally, in an expanded, futuristic, and forward-looking description, we discuss the different ML approaches, varying from artificial neural networks and vector regression to Bayesian and multi-objective optimization methods, suited for enabling ink formulation and process optimization (associated with the handling of the ink) that ensure inks that result in printed traces with desired conductivity, minimum voids and cracks, and arrested coffee stain effect. Given the surge of ML and artificial intelligence (AI) approaches in upending our technological and engineering enterprise and the desperate need to optimize the ink quality for targeted printed electronics applications, we believe that this is an extremely timely perspective article.

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