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Study on Factors Affecting the Properties of Conductive Ink Prepared from Graphite and Polyvinylpyrrolidone

Nguyen Thanh Binh[1,*] , Nguyen Thi Mo[2]

[1] *Faculty of Chemistry, College of Chemical Defense Officer, Chemical Corps*
[2] *Faculty of Chemistry, VNU University of Science, Vietnam National University, Hanoi*
* *Email: binhnbc1982@gmail.com*

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ABSTRACT

The main components of the conductive ink in this study are graphite and polyvinylpyrrolidone (PVP), with their ratio significantly influencing the ink's rheological and electrical properties. This research investigates the effect of PVP concentration on conductivity, film adhesion, and ink flexibility; and the influence of graphite content on the ink's conductivity. The results showed that a PVP concentration of 5% and a graphite content of 0.5 g/mL yielded optimal viscosity, adhesion, and electrical conductivity. Moreover, the incorporation of carbon black (CB) into the ink formulation considerably improved its conductivity.

Introduction

In recent years, conductive inks have emerged as a promising development in electronics. These inks typically contain conductive metallic nanoparticles such as copper, silver, or gold, or nanostructured carbon materials, dispersed in a polymer matrix with a solvent to form a printable liquid suitable for various substrates [1,2,5,6]. Conductive inks are highly applicable in many smart electronic devices including biosensors, flexible electronics on plastic or paper substrates, energy storage devices like batteries and capacitors, and semiconductors [3,7,10,12]. They are low cost, lightweight, and ease of fabrication offer significant advantages over traditional conductive materials [10]. Consequently, conductive inks have attracted substantial research interest globally. The number of studies aiming to develop novel conductive ink formulations has increased markedly in recent years. Common materials used include metal particles (silver, gold, copper), carbon-based materials (graphite, graphene, carbon nanotubes), conductive polymers (e.g., poly(3,4-ethylenedioxythiophene), polyaniline), or hybrid systems combining multiple components (e.g., graphene/silver, CNTs/polymer) [7,8,9]. Depending on the intended application, different ink types are selected. Conductive inks based on graphite and PVP are of particular interest due to their low cost, ease of fabrication, and promising application potential [4,10,11].

In this study, we focus on examining the factors influencing the properties of conductive inks formulated from graphite and polyvinylpyrrolidone (PVP). Graphite serves as the primary conductive material, while PVP functions as a stabilizer, dispersant, and binder, ensuring good dispersion of graphite particles and forming a flexible film on the printing substrate. Graphite–PVP-based conductive inks are low-cost and exhibit stable electrical conductivity, which makes them promising for applications in sensors and printed electronic circuits [13,14,15]. In this work, we further investigate the effect of incorporating black carbon (BC) into graphite–PVP inks and compare its influence on the electrical conductivity.

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Experimental

Materials

Hydrochloric acid solution (HCl, 1 M), sodium chloride solution (NaCl, 1 M), and ethanol solution (3%) were employed as reagents. Polyvinylpyrrolidone (PVP, $(C_6H_9NO)_n$, K90, China) with a purity of 99% was used as the polymer stabilizer. Commercial graphite (GTM, China) with a particle size of 1–20 µm and a purity above 99% was applied as the main conductive filler. Black carbon (BC, China) with a purity above 99% and a particle size of 10–80 nm was also utilized.

Methods

Viscosity Measurement: Following Vietnamese Standard TCVN 3171.

Electrical Resistance Measurement: Following Vietnamese Standard TCVN 10530:2014.

Ink Preparation Procedure: The ink was prepared by dissolving PVP in a 1:2 (ethanol:water, by weight) solvent mixture using mechanical stirring. PVP was gradually added to avoid clumping, and stirring continued for 15 minutes at 1000 rpm until a homogeneous, clear solution was obtained. Graphite and CB were weighed accurately and dispersed into the PVP solution at 1500 rpm for 30 minutes. The final ink was then characterized for viscosity and electrical resistance.

Results and discussion

Effect of PVP concentration on conductive ink properties

Electrical conductivity

Graphite-based inks were prepared with a constant graphite content (0.5 g/mL) and varying PVP concentrations (1÷7%). The inks were printed on different substrates: hydrophilic (paper, polyethylene - PE) and hydrophobic (polyvinyl chlorua- PVC). Table 1 summarizes the results:

Table 1. Electrical resistance of GTM inks with varying PVP concentrations

PVP (%)		1	3	5	7
Viscosity (mPa.s)		130	450	1161	Over
Resistance (KΩ)	PVC	4.34	2.40	3.82	4.11
	Paper	8.54	7.41	5.30	9.89
	PE	4.50	2.60	3.70	4.00

The results indicate that PVP concentration significantly affects both the electrical conductivity and rheological properties of the ink. Due to the high molecular weight of PVP, even at low concentrations, the ink solution becomes viscous. The viscosity of the ink increases proportionally with PVP concentration. However, the influence of PVP concentration on electrical conductivity is not linear. Initially, electrical resistance tends to decrease as PVP concentration increases from 1% to 3%, but then increases again at higher concentrations. This suggests that at low concentrations (e.g., 1%), the solution's viscosity is insufficient to create an environment that enables uniform dispersion and contact between conductive particles. Conversely, at higher PVP concentrations, the dense polymer binder matrix surrounding the graphite particles hinders electron mobility, increasing resistance. Therefore, the optimal PVP concentration lies in the range of 3–5%. Moreover, the effect of PVP concentration also varies depending on the substrate used for printing. The results show that inks printed on PVC and PE surfaces exhibit lower resistance compared to those printed on paper. This suggests that conductivity on paper is inferior due to its porous and absorbent nature, which allows graphite particles to penetrate into the substrate, reducing surface conductivity. In contrast, PVC and PET surfaces are smooth and non-absorbent, leading to a higher amount of graphite remaining on the surface and thus better conductivity. Consequently, the findings suggest that plastic substrates such as PVC and PE are more suitable for subsequent investigations.

Effect of PVP Concentration on Ink Film Durability

For printed inks, one of the key indicators of durability is the adhesion of the ink to the substrate surface. In graphite/PVP-based conductive inks, adhesion is primarily governed by the PVP component. The adhesion performance on a PVC substrate was evaluated using a tape-peel test, in which the printed ink film was peeled off using adhesive tape. The electrical resistance of the printed line was measured both before and after the peeling process. The results are presented in Table 2 and Figure 1.

Table 2. Conductivity results of ink prepared with various PVP concentrations (1% to 7%) and 0.5 g/mL graphite after a single tape-peel test.

PVP (%)	Resistance (KΩ) Initial	After tape removal
1	4.34	No signal
3	2.4	20.5
5	3.82	6.9
7	4.1	5.6

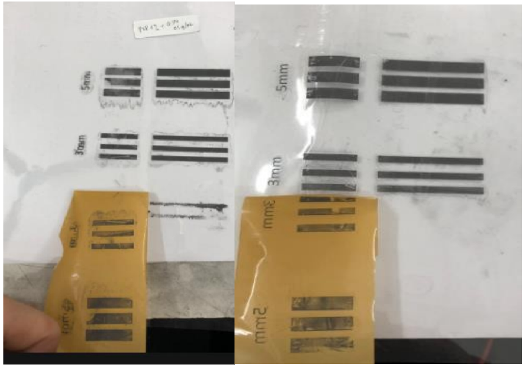

Figure 1. Images of conductive ink with 1% PVP (a) and 3% PVP (b) after a single tape-peel test

The results from Table 2 show that in the case of 1% PVP, the ink layer on the PVC substrate was almost entirely peeled off after the tape test, resulting in no detectable electrical signal. For the sample with 3% PVP, the resistance increased nearly tenfold compared to its initial value. In contrast, the 5% PVP sample exhibited only a twofold increase in resistance, while the 7% PVP sample showed a minimal increase of approximately 1.36 times. These findings indicate that ink adhesion improves with increasing PVP concentration. At 1%, the low PVP content is insufficient to bind the graphite particles and anchor them effectively to the PVC surface. Therefore, PVP concentrations of 3% and 5% offer better adhesion performance. Although the 7% PVP sample showed the least degradation in conductivity after peeling, its significantly higher viscosity renders the ink unsuitable for screen-printing applications.

Effect of PVP concentration on the flexibility of the ink

Based on the results of PVP concentration's influence on electrical conductivity and film adhesion, a PVP concentration of 5% was found to be the most suitable for screen-printing applications. The mechanical flexibility of the ink was evaluated by bending the printed ink lines over cylinders with different diameters. Three bending diameters were tested: 3.4 cm, 1.4 cm, and 1.1 cm.

Table 3. Electrical conductivity results of ink formulated with 5% PVP and 0.5 g/mL GTM graphite under different bending conditions.

Bending diameter	Flat surface	1.1 cm	1.4 cm	3.4 cm
Resistance kΩ/cm)	3.82	6.31	5.91	4.67

After the bending durability tests using cylinders of different diameters, the electrical resistance of the ink was observed to increase compared to its initial value. This increase in resistance is attributed to microcracks forming on the ink film when bent along a curved surface, which disrupts the conductive pathways [2]. However, the rise in resistance is relatively minor and does not compromise the overall conductivity of the ink. These results indicate that at a concentration of 5% PVP, the ink maintains good electrical conductivity and orms strong bonds with the conductive graphite particles.

Effect of Graphite content on the electrical conductivity of the ink

Graphite content significantly affects not only the electrical conductivity but also the processability (viscosity) of the ink. With the PVP concentration fixed at 5%, the influence of varying graphite content on the rheological properties and electrical resistance of the conductive ink was investigated. The results are presented in Table 4.

Table 4. Electrical resistance and viscosity of conductive ink with varying commercial graphite

Graphite Content (g/mL)	0.3	0.5	0.7
Viscosity (mPa·s)	450	1161	Over
Resistance (kΩ/cm)	3.22	2.27	4.10

Note: "Over" indicates viscosity exceeded the measurable limit of the instrument.

The results show that the sample with a graphite content of 0.5 g/mL exhibits lower electrical resistance compared to the sample containing 0.3 g/mL. This indicates that a higher graphite content leads to better conductivity, as a greater concentration of conductive particles on the surface facilitates more efficient electron transport [4]. However, it can also be observed that at excessively high graphite content (e.g., 0.7 g/mL), the ink loses its flowability, thereby hindering the printability and processing of the ink.

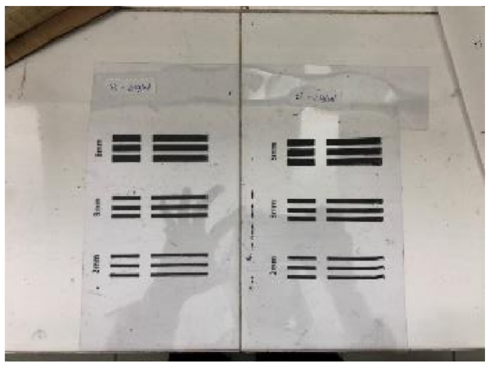

Figure 2. Image of graphite-based ink printed on a PVC substrate

Previous studies have shown that carbon black (CB) is often used alongside graphite in ink formulations to

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enhance electrical conductivity and increase surface area, thereby improving electron transport [3]. Thaís Cristina de Oliveira Candido and colleagues [3] investigated the effect of the graphite/carbon black ratio on the electrochemical properties of the ink. Their results indicated that at low CB content, the ink exhibited poor conductivity; specifically, a composition of 37.5% graphite and 13.5% CB resulted in optimal electrochemical performance and excellent adhesion. However, when the ratio was adjusted to 35% graphite and 15% CB, the electrochemical performance declined, and the ink showed poorer adhesion. This outcome may be related to the increased amount of CB compared to previous ratios, as the higher mass of CB may hinder ink adhesion. To improve the conductivity of graphite ink in this study, CB was incorporated into the formulation at a GMT/CB weight ratio of 7:3. The results showed a significant improvement in electrical conductivity, with the ink's resistance decreasing by nearly five times.

The electrical resistance of the ink significantly decreased after the incorporation of carbon black (CB), dropping from 2.27 KΩ for the sample with only graphite (GMT) to 0.50 KΩ for the sample with a GMT/CB ratio of 7/3. The enhancement in electrical conductivity upon the addition of BC can be attributed to its nanoscale particle size, which is significantly smaller than that of graphite. This enables better dispersion across the surface and the formation of conductive bridges between graphite particles, resulting in a continuous electron conduction network with improved stability. The ultrafine BC particles also interact with PVP, producing a more uniform ink with good elasticity, resistance to delamination under bending, and strong adhesion to the printing substrate. These findings indicate that the incorporation of BC markedly improves electron transport in graphite-based inks, thereby enhancing their overall electrical conductivity.

Conclusions

Based on PVP concentration and commercial graphite content evaluation with respect to adhesion strength, film durability, flexibility, and electrical conductivity of the printed ink, the optimal formulation was determined to be 5% PVP and 0.5 g/mL graphite. Notably, adding of CB to the PVP and GMT system significantly enhanced the ink's electrical conductivity-improving it by a factor of five compared to the formulation containing only 5% PVP and 0.5 g/mL GMT.

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