Developing Conductive Ag and Carbon Pastes for Screen Printing Applications

Gizem ŞAHİN[1]
Assoc. Prof. Dr. Mustafa ŞEN[2]
[1,2]**Biomedical Engineering Department, İzmir Katip Çelebi University, İzmir, Türkiye**
[2]**ORCID: 0000-0002-2421-9184**

ABSTRACT

The development of electrochemical devices has become a significant focus in the field of biomedical engineering in recent years. Particularly, there has been an increased interest in low-cost and disposable solutions, as they overcome the challenges associated with the continuous renewal of traditional electrodes and reduce sample volume. Disposable electrochemical devices offer practical solutions in areas such as health monitoring, environmental monitoring, and food analysis, providing user-friendly approaches. This study focuses on optimizing screen-printed electrodes (SPEs) through the development of Ag and carbon-based conductive pastes. Silver paste synthesized using silver nanoparticles (AgNPs) exhibited high conductivity suitable for electrochemical applications. Characterization tests, including electrical resistance measurements and rheological analysis, confirmed the paste's potential for thin film electrode production. Carbon-based pastes, incorporating multi-walled carbon nanotubes (MWCNTs) and activated carbon, were also evaluated for their electrical properties and substrate adhesion. Microstructural analysis using SEM and FTIR spectroscopy validated the morphology and surface chemistry of AgNPs synthesized, while X-ray diffraction (XRD) analysis confirmed their crystallographic structure. Overall, this study contributes valuable insights into the fabrication and optimization of conductive pastes for screen-printed electrodes, paving the way for their effective utilization in biomedical, environmental, and food monitoring applications.

Keywords: Screen-printed electrodes, Conductive pastes, AgNPs, MWCNT, Electrochemical sensors

1. INTRODUCTION

In the past decade, electrochemical devices have been developed as an up-and-coming biomedical engineering focus area. Interest in low cost and disposable solutions has increased, for they overcome the challenges of continuous renewal of traditional electrodes and reduce sample volume (Hayat et al.,2014, Şen et al. 2023, Aydin et al. 2017, Seven et al. 2020). Electrochemical

disposable devices represent a very practical solution for health and environmental monitoring, food analysis, etc. and have been translated into user-friendly approaches (Şen et al. 2022, Oğuz et al. 2024, Şen et al. 2024). Screen printed electrodes represent a very popular electrode type within the electrochemical sensing applications field. They are preferred because of their high speed of production, low cost, repeatability, and the possibility of large-scale manufacture (Henrique et al., 2021). Screen-printed electrodes prepared with commercial conductive inks can be optimized by adding extra components such as nanoparticles, conductive polymers, and carbon-based materials to enhance their performance (Ibáñez-Redín et al.2018). Normally, conductive inks are composed of a polymer base, conductive material, and solvents—the choice and ratio of which would be very important considering electrochemical performance and sensor stability. Among them, water-based conductive inks have attracted great attention due to their low cost, eco-friendliness, and convenient processing. Carbon materials and metallic nanoparticles are largely sought-after conductive components to enhance screen-printed electrodes. Conductive inks used for constructing and repairing electrical circuits have a significant role in the development of disposable electrochemical sensors today. Actually, their commercialization process is underway while labs continue developing new strategies which can be prepared at laboratory conditions so as to minimize costs. Research is being worked on different methodologies and materials which formulate conductive inks with an application to the fabrication of sensors and biosensors (Camargo et al. 2022). In such respect, AgNPs are shown as one important component in screen-printed electrodes, showing very high conductivity and antioxidant property and enough stability at temperatures (Carvalho et al. 2021). Most of the time, Ag-based inks are preferred since they stick better to substrates in comparison. This study aims to develop and characterize conductive Ag and multi-walled carbon nanotubes (MWCNT)-based carbon pastes suitable for the fabrication of screen-printed electrodes (SPEs). MWCNTs are best known for their exceptional mechanical, electrical, and thermal properties, making them invaluable in various scientific and industrial

applications. They have found widespread applications across diverse research fields, ranging from creating highly efficient conductive tracks for electronic devices to developing advanced conductive pastes for sensors (Sikora et al. 2015).

2. MATERIALS AND METHODS

2.1. Materials

MWCNTs (Nanografi, Turkey), carboxyl-functionalized MWCNTs (MWCNT-COOH) (Nanografi, Turkey), acrylic varnish (Dyo, Turkey), varnish hardener (Dyo, Turkey), liquid paraffin (Sigma-Alrich, USA), 14% polystyrene-block-polystyrene copolymer (Sigma-Alrich, USA), toluene solvent (Sigma-Alrich, USA), silver nitrate ($AgNO_3$) (Sigma-Alrich, USA), citric acid (Sigma-Alrich, USA), iron (II) sulfate heptahydrate ($FeSO_4 \bullet H_2O$) (Sigma-Alrich, USA), and ethanol (Sigma-Alrich, USA).

Table 1. Combinations indicating ratios to be tested in the production of carbon-based conductive pastes.

	1	2	3	4	5	6
Carbon Material	% 5	% 5	% 10	% 10	% 20	% 20
Acrylic Varnish	% 90	% 85	% 85	% 80	% 75	% 70
Varnish Hardener	% 5	% 10	% 5	% 10	% 5	% 10

2.2. Conductive Ag paste

To prepare the Ag nanoparticles (AgNPs), 1 g of $AgNO_3$ was initially mixed with 70 mg of $C_6H_8O_6$ in 50 mL of dH_2O. The mixture was sonicated for 30 minutes to homogenize it. Concurrently, 4 g of $FeSO_4$ powder was dissolved in 50 mL of dH_2O. The $AgNO_3$ and $FeSO_4$ solutions were then combined and sonicated for 2 hours to complete the synthesis of AgNPs. Upon completion, the AgNPs were centrifuged at 5000 rpm for 10 minutes to precipitate the nanostructures. The supernatant was carefully removed, and the precipitate was cleaned and washed three times with

an equal amount of water to achieve the desired properties. This washing process was repeated three times using ethanol. For the production of Ag paste from AgNPs, 1 gram of polystyrene was dissolved in 5 mL of toluene to prepare a homogeneous solution. To this solution, 88 µL of polystyrene-toluene solution was added and mixed with 40 mg of AgNPs for 5 minutes using a probe sonicator. This process resulted in the production of Ag paste.

2.3. Conductive carbon paste

Carbon-based conductive pastes required for the production of SPEs were prepared as part of this study. The materials used in the preparation included multi-walled carbon nanotubes (MWCNTs), carboxyl-functionalized MWCNTs (MWCNT-COOH), and activated carbon. For each type of carbon material, a mixture of liquid varnish, carbon material, and paraffin was prepared in specific proportions, as shown in Table 1. The pastes were manually mixed to a paste-like consistency for 3-5 minutes, a process repeated for six different concentrations. The conductive pastes were then applied onto acetate substrate surfaces and allowed to dry for one day. After drying, resistance measurements were performed using a voltmeter with a 1 cm gap between the probes.

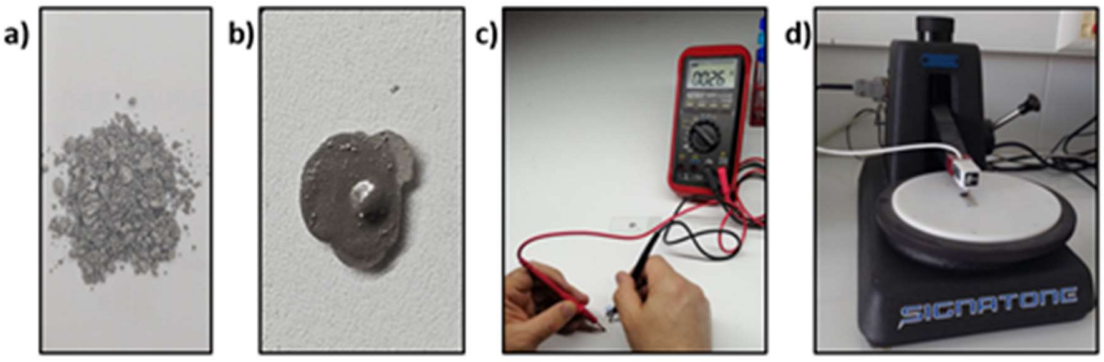

Fig. 1. Images of synthesized AgNPs (a) and Ag paste (b). The resistance of the Ag paste was measured separately using a multimeter (c) and a four-point electrical measurement device (d).

3. RESULTS AND DISCUSSION

This work investigated the electrical and physical properties of synthesized Ag paste. As can be seen in Fig. 1a and b, first the AgNPs and then the conductive Ag paste was successfully produced

using the given methodology. The initial resistance measurement showed a low value of 2.6 Ω, indicating high conductivity (Fig. 1c). Electrodes printed with the Ag paste, about 5 μm thick, had a resistivity of $10.9 \pm 0.1 \times 10^{-5}$ Ω·cm using a four-point probe (Fig. 1d). These results confirm the Ag paste's suitability for thin-film production. Rheometer tests identified the viscosity characteristics of the Ag paste. The "zero-rate viscosity" was 334.433 Pa·s for undiluted Ag paste and 102.980 Pa·s for ethanol-diluted paste (1:10 ratio), indicating adjustable viscosity for different purposes. SEM analysis (Fig. 2a) revealed that the Ag nanoparticles had flake-like, homogeneous structures, confirming the quality and efficacy of the synthesis method. FTIR spectroscopy showed hydrogen-bonded groups on the AgNPs surface, with bands at 3434 cm^{-1} (alcohol, phenol, or water) and 2922 cm^{-1} and 2856 cm^{-1} (aliphatic C–H groups), indicating organic component interactions. The presence of a carboxyl group at 1626 cm cm^{-1} was also confirmed (Fig. 2b). XRD analysis revealed crystallographic peaks at 2θ angles of 38º, 44º, 64º, 77º, and 82º, consistent with silver crystals (111, 200, 220, 311, 331) per PXRD standards (Ref. No. 01-087-0718) and JCPDS Card No. 87-0597 (Fig. 2c). In summary, these results demonstrate the successful synthesis and characterization of Ag paste, highlighting its potential for versatile applications.

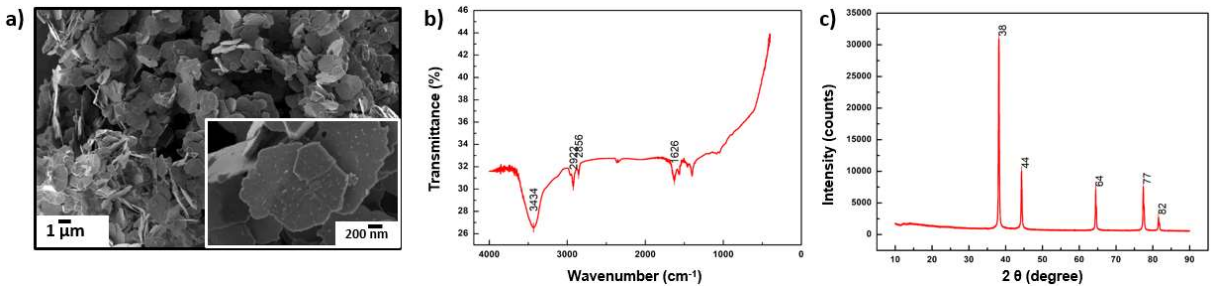

Fig. 2. SEM image of AgNPs (a), FT-IR spectrum (b), and XRD results (c).

The carbon paste combinations were applied onto an acetate substrate and allowed to dry. The dried conductive pastes were then examined. As shown in Fig. 3, the pastes prepared with MWCNT and MWCNT-COOH in columns 5 and 6 did not adhere well to the acetate substrate and fragmented. It was observed that as the MWCNT content increased, the resistance of mixtures

1, 2, 3, and 4 decreased from 400 Ω to 50 Ω. All pastes were tested for screen-printed electrode (SPE) production. Four-point electrical measurements were conducted for the MWCNT/SPE prepared from mixture 3, which showed a layer resistance of $593.8 \pm 99.3 \times 10^{-3}$ Ω/sq. The lowest resistance for MWCNT-COOH pastes was 2 Ω for mixture 4, which was then used for SPE production. The resistance of activated carbon pastes ranged from 5 kΩ to 140 kΩ. Mixture 6, with the lowest resistance, was not preferred for SPE production due to its high viscosity. Therefore, mixture 5, with a resistance of 7.8 kΩ, was chosen. The results highlight the effects of varying MWCNT and MWCNT-COOH concentrations on the electrical properties of pastes. Selecting appropriate paste formulations with specific resistance characteristics is crucial for the successful fabrication of SPEs and their performance in various applications.

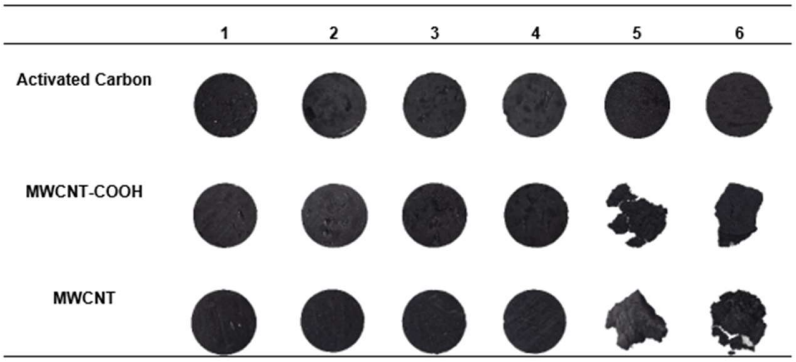

Fig. 3. Images of the produced carbon-based conductive pasta on acetate substrate.

4. CONCLUSION

This work focuses on developing and characterizing conductive pastes for screen-printed electrodes (SPEs) to optimize electrical performance for electrochemical sensing. Silver nanoparticle-based paste showed excellent conductivity with low resistance and suitable viscosity for various applications. Carbon-based pastes, using MWCNTs, carboxyl-functionalized MWCNTs, and activated carbon, exhibited varying resistance levels. Optimal formulations were identified despite challenges in viscosity and adhesion. The study underscores the importance of

paste formulation for achieving desired SPE properties, enhancing sensor performance, sensitivity, stability, and cost-effectiveness. Future research will explore further optimization and new materials for disposable electrochemical sensors. This research offers valuable insights into conductive pastes for SPEs, facilitating their use in biomedical, environmental, and food monitoring applications.

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