Inkjet-Printed Flexible Temperature Sensor using Hybrid Reduced Graphene Oxide – Silver Nanoparticles (rGO/AgNPs) Conductive Ink and Silver Nanoparticles Ink

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Abstract. This paper presents a new temperature sensor, inkjet-printed with the in-house developed hybrid rGO/Ag nanoparticles ink (rGO/AgNPs ink). Its performance is studied, and the results indicate that its sensitivity is better than the commercial sensor. The meander—shaped electrodes were fabricated using drop-on-demand inkjet printer (Fujifilm Dimatix 2850 printer) on polyethylene terephthalate (PET) substrate. Compared to the sensitivity 0.0543 Ω/°c of the sensor developed with commercial ink, the in-house developed sensor shows higher sensitivity 0.1086 Ω/°c. Besides, the printed sensors exhibit its resistance increasing linearly with temperature from 30°C to 100°C. The bending tests results also prove that the characteristics of the sensors do not vary significantly, indicating excellent mechanical stability and flexibility. Therefore, the flexible inkjet-printed temperature sensor with the in-house hybrid rGO/AgNPs ink is recommended for the large-scale productions and implementations.

1. Introduction
Unlike the conventional technologies, printed electronics today offers an alternative with many unique advantages and used in various areas. For example, in energy storage, flexible devices and health applications. Inkjet-printing based fabrication has attracted immense attention among many available manufacturing techniques. Its attractions come from the low-cost manufacturing, eco-friendly fabrication technologies, and the mechanical flexibility and lightweight printed devices.

In inkjet-printing methods, sensing materials is one of the keys that can generate many possible results to one solution. On the other hand, the sensing materials significantly relies on the ink type and ink quality. For instance, current existing works [1] and [2] have demonstrated that impressive results are made with copper (Cu) and gold (Au) inks. Compared to these inks, the hybrid reduced-Graphene Oxide/Silver nanoparticles ink (AgNPs) is more preferred due to AgNPs have lower reactivity to air [3], lower resistance to corrosion [4], lower melting temperature [5] and lower cost [6]. Nonetheless, its cost is still the greatest concern to the industrial interest when the production is at large-scale.
Hence, in-house hybrid rGO/AgNPs ink has been purposed, developed, and presented by MIMOS berhad currently. The performance of the sensor printed with commercial silver nanoparticles ink is studied and compared with the performance of the in-house flexible inkjet-printed temperature sensor developed with hybrid rGO/AgNPs ink. Similar to [10], the investigation of the printing properties on the PET using the in-house hybrid rGO/Ag ink have also been carried out. Results have indicated that the sensitivity of the in-house temperature sensor is very close to the commercial sensor.

2. Materials and Methods

2.1. Sensor Design
Meander-shaped electrode are designed and built with Pattern Editor on DMP program. Once the pattern is created, it is saved in pattern file (.ptn) which allows easier in modifying the pattern such as changing the resolution and the number of printing layers based on the requirement. Table 1 below shows the geometry parameter of the electrode while Figure 1 illustrates the designs and specifications of electrode.

![Figure 1: Meander-shape electrode](image)

Table 1: Meander-shape electrode design parameter

| Design parameter | Value (mm) |
|------------------|------------|
| Width, W         | 0.5        |
| Gap, G           | 0.4        |
| Padding, P       | 1.5        |
| Total width, W<sub>T</sub> | 6.9 |
| Total length, L<sub>T</sub> | 13.2 |

2.2. Inks and Substrate Materials preparations
Two types of nanoparticles ink were selected in this research which are the commercial AgNPs inks and the in-house rGO/AgNPs ink. The commercial AgNPs ink used is obtained from Novacentrix® (Metalon® JS-B25HV). It is conductive Nanosilver ink designed to produce patterns/circuits on porous/non-porous substrates such as PET, inkjet papers and glass. This AgNPs ink was specially formulated for the Fujifilm DMP printer and the waveform for DMP printheads is provided. Table 2 shows the ink properties of Metalon® JS-B25HV.

Table 2: Technical specifications for commercial AgNPs ink

| Properties               | Value |
|--------------------------|-------|
| PH                       | 6.0-8.0 |
| Viscosity (cP)           | 8-10  |
| Surface tension(dyne/cm) | 30-32 |
| Particle size (nm)       | 60-80 |
Secondly, it is the hybrid rGO/Ag nanoparticles ink (rGO/AgNPs ink). It is developed internally by MIMOS researchers. This conductive nanoparticle ink is specially formulated for higher compatibility with Fujifilm DMP printer. Table 3 illustrates the ink properties of in-house hybrid rGO/AgNPs ink.

| Properties             | Value       |
|------------------------|-------------|
| PH                     | 7.5-8.5     |
| Viscosity (cP)         | \(\leq 10\) |
| Surface tension(dyne/cm) | 38-39     |
| Particle size (nm)     | 290-900     |

The substrate in this research is Polyethylene terephthalate (PET) which obtained from AgIC. The PET is treated with transparent film with a thickness of 107 \(\mu\)m \(\pm\) 32 \(\mu\)m. The treated layer allows conductive ink to dry and become conductive quickly without any pre/post processing. The PET comes in A4 size (210 mm * 297 mm) and allow users to cut into any size based on the different requirement.

2.3. Inkjet Printing Process

The temperature sensor was fabricated on PET substrate with Fujifilm Dimatix printer (model: DMP 2850) inkjet printer which is illustrated in Figure 2. It is a drop-on demand-demand (DOD) printer using the piezoelectric inkjet technology where the droplets are ejected only when required. The DMP printers are compatible with most solvents because they allow users to customize their own inkjet printability parameters to suit their fluids/inks to get the optimum printing performances. This printer offers two types of disposable print cartridges which are 1 pL (DMC-11601) and 10 pL (DMC-11610). In this research, 10 pL disposable print cartridges which contain 16 nozzles that are linearly spaced at 254 \(\mu\)m spacing is selected. This cartridge can hold 0.2 ml up to 1.5 ml volume of ink to minimize waste.

Before filling ink into the cartridge, the ink is required to be degas approximately 15 minutes to remove the dissolved gas. Degassing is useful to improve jetting performance for most fluids/inks. The fluid (~1.5 ml) is filled into the cartridge using syringe. Then install syringe filter and needle, insert into fluid module fill port and inject the fluid/ink. Lastly, attach the fluid module to the jetting module (Figure 3). The syringe filter is used to remove the large particles/aggregates to prevent the nozzle head clog. In general, the particles in the fluid/ink should be 1/100\(^{\circ}\) of the size of nozzle. The effective diameter of nozzle (DMC-11610) is approximately 21 \(\mu\)m, so the 0.2 \(\mu\)m filter is chosen in this research. It is recommended by the manufacturer to load the cartridge into the printer immediately and keep the cartridge at least 30 minutes in idle state.
The printing quality of the pattern are affected by various inkjet printability parameters. To obtain high printing quality, the printing parameters are required to be optimized. The inkjet printability parameters that are investigated include:

- Substrate thickness (um)
- Tickle Control (kHz)
- Stage temperature (°c)
- Cartridge temperature (°c)
- Jetting voltage (v)
- Meniscus Setpoint (inches H₂O)
- Cartridge Print Height (mm)
- Number of jets to use
- Leader bar
- Resolution (dpi)
- Number of printing layer

The optimal inkjet printability parameters that were set in DMP software for both commercial AgNPs ink and the in-house hybrid rGO/AgNPs ink were illustrated in Table 4.

Table 4: Inkjet printability parameters for commercial AgNPs ink and in-house hybrid rGO/AgNPs ink

| Printer Parameter                  | JS-B25HV | Hybrid rGO/AgNPs |
|------------------------------------|----------|------------------|
| Substrate Thickness (um)           | 200      | 200              |
| Tickle Control (kHz)               | 5kHz     | 5kHz             |
| Stage Temperature (°c)             | 40       | 40               |
| Cartridge Temperature (°c)         | 30       | 30               |
| Jetting Voltage (v)                | 22       | 15               |
| Meniscus Setpoint (inches H₂O)     | 3.5      | 4                |
| Cartridge Print Height (mm)        | 0.5      | 0.5              |
| Number of jets to use              | 1        | 1                |
| Leader Bar                         | Enable   | Enable           |
| Resolution (dpi)                   | 1270     | 1270             |
| Number pf printing layer           | 1        | 1                |

Beside the inkjet printability parameters, cleaning cycle is significantly required to help in improving the printing quality by avoiding the nozzles clogging. The cartridge nozzle heads before, during and after printing is important to keep clean for clean nozzles. Before printing and at every 20 cycles during printing, the nozzle was cleaned by the printer with purge function. The function of the purge is to push the ink/fluid out through the nozzle with high pressure to eliminate the air.
2.4. Sensor Characterization Techniques
The geometrical and morphology characteristics was studied with Olympus microscope and the fiducial camera of printer. Microscope was utilized to obtain the sensor dimension and to observe the morphology of the sensor. The dimension of the sensor is subsequently measured using the Olympus software and the design pattern specification was investigated.

The sensors were placed onto a high accuracy hotplate. The sensors were characterized by connected to Fluke digital multimeter to measure the resistance value at the temperature from 30°C to 100°C with 10°C increment.

In order to evaluate the sensors flexibility, bending test was carried out with difference diameter of curvature. The resistance values were recorded using multimeter and studied.

3. Results and Discussion

3.1. Geometrical and Morphology Characterization
Figure 4 and Figure 5 depict the microscopic image for both commercial AgNPs ink and in-house hybrid rGO/AgNPs ink. The printing quality for both inks were good. This is because sharp edges, clean and uniform ink deposition were observed. Table 5 shows dimension differences between the design pattern specification and the actual printed sensor. Based on the results, the difference between design and actual printed is found less than 10% (commercial AgNPs ink) and less than 5% (in-house hybrid rGO/AgNPs ink).

![Microscopic image for commercial AgNPs ink](image1)

![Microscopic image for in-house hybrid rGO/AgNPs ink](image2)
Table 5: Dimension difference between design pattern specification and actual print sensor for both inks

| Design Parameter | Design AgNPs ink | Commercial AgNPs ink | Hybrid rGO/AgNPs ink |
|------------------|------------------|----------------------|----------------------|
| Width (mm)       | 0.5              | 0.5±0.04             | 0.5±0.02             |
| Gap (mm)         | 0.4              | 0.4±0.04             | 0.4±0.02             |
| Pad (mm)         | 1.5              | 1.5±0.03             | 1.5±0.02             |

3.2. Electrical Characterization

The temperature sensors were electrically characterized with a Fluke multimeter. The temperature varied from 30°C to 100°C, with 10°C increment in each step. Figure 5 and Figure 6 depict the temperature-electrical resistance relationship for both commercial AgNPs ink and in-house hybrid rGO/AgNPs ink. Total of six printed sensors (3 set for each ink types) were selected. The results showed a linear relationship between resistance and temperature. The coefficients of linearity after linear fitting were 0.9873 (AgNPs ink) and 0.9898 (hybrid rGO/AgNPs ink).

![Sensor 1](image1)

\[ y = 0.0488x + 40.402 \]

\[ R^2 = 0.9873 \]

![Sensor 2](image2)

\[ y = 0.0443x + 38.221 \]

\[ R^2 = 0.9948 \]
Figure 6: Resistance vs temperature for commercial AgNPs ink

**Sensor 3**

\[ y = 0.0563x + 38.702 \]

\[ R^2 = 0.988 \]

**Sensor 4**

\[ y = 0.2545x + 162.41 \]

\[ R^2 = 0.9898 \]

**Sensor 5**

\[ y = 0.1385x + 97.788 \]

\[ R^2 = 0.9907 \]
Figure 7: Resistance vs temperature for in-house hybrid rGO/AgNPs ink

The relationship between resistance and temperature change can be calculated by

$$\frac{R_T - R_0}{R_0} = \alpha \Delta T$$  \(1\)

Where \(R_T\) represents the resistance value at temperature \(T\) and \(R_0\) is the resistance value at temperature 30\(^\circ\)C, \(\Delta T\) is the variation in temperature. \(\alpha\) is known as the temperature coefficient of resistance (TCR) which indicates the resistance change factor per degree Celsius of temperature change.

$$\alpha = \frac{R_T - R_0}{R_0(\Delta T)}$$  \(2\)

Besides that, the sensitivity (S) of the sensors can be obtained from

$$S = \frac{R_T - R_0}{\Delta T}$$  \(3\)

Table 6 shows the TCR and sensitivity of the printed for AgNPs ink and in-house hybrid rGO/AgNPs ink. From the table, the experiment 5 which printed the sensors with hybrid rGO/AgNPs ink produced the highest sensitivity of 0.1086 \(\Omega/\)°C.

Table 6: Sensitivity and TCR of printed temperature sensor

| Sensor | Temperature coefficient of resistance(°C\(^{-1}\)) | Sensitivity (\(\Omega/\)°C) |
|--------|-----------------------------------------------|---------------------------|
| 1      | 12.3626 \(\times 10^{-4}\)                    | 0.0514                    |
| 2      | 11.6026 \(\times 10^{-4}\)                    | 0.0457                    |
| 3      | 13.4704 \(\times 10^{-4}\)                    | 0.0543                    |
| 4      | 9.9668 \(\times 10^{-4}\)                     | 0.1071                    |
| 5      | 10.0343 \(\times 10^{-4}\)                    | 0.1086                    |
| 6      | 7.3363 \(\times 10^{-4}\)                     | 0.0786                    |

3.3. Flexibility test

Bending test was conducted to study the bending effects on the sensor resistance. The printed sensors were bend around 20 mm diameter curvature. The bending cycle was set to 5 at room temperature. The table 6 shows the change in resistance before and after bending test. After the bending test, the resistance of printed sensors increased 0.2 \(\Omega\) - 0.6 \(\Omega\) which is around \(\pm 0.6\%\) increment.
Table 7: Resistance value for bending test

| Sensor | Initial resistance value (Ω) | Resistance value (Ω) after bending test | Change in resistance (Ω) |
|--------|-----------------------------|------------------------------------------|-------------------------|
| 1      | 41.6                        | 41.9                                     | +0.3                    |
| 2      | 39.4                        | 39.9                                     | +0.5                    |
| 3      | 40.3                        | 40.5                                     | +0.2                    |
| 4      | 170.1                       | 170.3                                    | +0.2                    |
| 5      | 101.2                       | 101.8                                    | +0.6                    |
| 6      | 172.8                       | 173.1                                    | +0.3                    |

4. Conclusion
DOD inkjet printer (DMP2850) is used to fabricate flexible inkjet-printed temperature sensors with two different kinds of inks which are the commercial AgNPs ink (JS-B25HV) and the in-house developed hybrid rGO/AgNPs ink. The sensitivity test is conducted within the temperature range of 30°C to 100°C. The resistance of both sensors increases linearly with higher temperature. The sensitivity of the sensor developed with commercial ink is 0.0543 Ω/°C, whereas the in-house developed sensor shows higher sensitivity 0.1086 Ω/°C. Moreover, the bending tests on the in-house developed sensor is found that the sensor resistance increased in 0.6%. In other words, the printed sensors demonstrate high stability under stress. Thus, the in-house developed sensor with in-house hybrid rGO/AgNPs ink is more recommended for the large-scale productions and implementations in industries compared to the sensor built with the commercial ink.

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Acknowledgments
The author wishes to thank Mrs Nor’azah Abdul Rashid and Dr. Lee Hing Wah for their technical support and guidance. A profuse thanks to my supervisor Dr. Huzein and Dr Haris for giving the guidance and kind support.