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## Review Article

# A Review on Electrical Conductivity of Cu–graphene Nanocomposites in terms of Production Methods and Reinforcement Ratios

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## ABSTRACT

Graphene is excellent material and is highly potent in terms of mechanical, electrical, optical, and thermal properties. Owing to these features, this material is used as a nanofiller for metal-based composites. Although many studies have focused on different attributes of graphene, the most remarkable is its electrical conductivity. In addition, copper, which exhibits one of the highest electrical conductivities among metal materials, is used in many different fields, especially in the electrical-electronics industry. Therefore, studies on the changes in the electrical properties of composites obtained using these two materials have expanded in recent years. In this study, the electrical properties of copper-graphene based nanocomposites produced using powder metallurgy are investigated. The changes in the electrical conductivity of the composites compared to the pure specimen are discussed in terms of graphene reinforcement and processing methods. The production methods and mixing techniques that achieve the most suitable electrical conductivity values have been comparatively evaluated. The graphene amount was considered in terms of production cost.



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## 1. Introduction

Research on materials has an extremely important place in the development of technology. Traditional solutions in many fields of science, such as nanotechnology, medicine, and engineering, are changing because of newly developed elements. By utilizing these technologies, composites with better properties than the materials that comprise their

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structural components can be produced by combining multiple substances (Janas & Liszka, 2017; Katiyaret al., 2021; Ranjan & Bajpai, 2021).

Among these superior ingredients, graphene-reinforced copper matrix composites constitute an important research topic. The superior temperature resistance, good corrosion resistance, and high conductivity seen in some studies make these materials stand out (Jamwal et al., 2020).

The high electrical and thermal conductivity of copper, which is the most important building block of copper–graphene nanocomposites, has come to the fore and become an indispensable element of the electronics industry. As a transition metal, Cu has a face-centered cubic crystal structure. It is solid at room temperature with a solid density of approximately 8.96 g/cm<sup>3</sup>. The most important mechanical properties of copper are its soft, ductile, and machinable nature. However, these features narrow the usage area because they cause plastic deformation (Almonti et al., 2022; Guo et al., 2007; Schlesinger et al., 2011; Varol et al., 2022; Zhang & Han, 2022)

Graphene, an allotrope of carbon, is a one-atom-thick, two-dimensional, honeycomb lattice material that can transmit electricity, heat, and light. Owing to these properties, it has been used in many fields of industry and science. The sigma bonds in graphene are strong. Therefore, while making significant contributions to the mechanical properties, this material also exhibits high heat dissipation. There are many derivatives of graphene (Gr), such as graphene oxide (GO), reduced graphene oxide (RGO), and graphene nanoplatelets (GNPs) (Ali et al., 2021; Hidalgo-Manrique et al., 2019; Kaiser, 2018; Murmu et al., 2022; Utpat & Kulkarni, 2022; Zhang et al., 2020)

Graphene, which is used in many different fields, has great potential for research applications (Elmgerbi et al., 2022; Jeganmohan et al., 2020; Murmu et al., 2022). The structural features of graphene are detailed in Table 1.

**Table 1.** Important properties of graphene (E.R.=±3) (Asgharzadeh & Eslami, 2019; Hansora & Mishra, 2017; Geim & Novoselov, 2007; Güler & Bağcı, 2020; Kumar & Xavior, 2014; Shao et al. 2020; Zhang, 2022)

| No | Properties of GNPs                         | Values               |
|----|--|----------------------|
| 1  | Purity Degree                              | (%) 99               |
| 2  | Mass (bulk) density (g/cm <sup>3</sup> )   | ~0,3                 |
| 3  | Real Density (g/cm <sup>3</sup> )          | 2,25                 |
| 4  | Thickness (nm)                             | ~1-2                 |
| 5  | Surface Area (m <sup>2</sup> /g)           | 2600                 |
| 6  | High-temperature resistance                | (-)75 / (+) 200 °C   |
| 7  | Thermal Conductivity (WK <sup>-1</sup> /m) | 4840-5300            |
| 8  | Electron Mobility cm <sup>2</sup> / (V.s)  | ~2,5×10 <sup>5</sup> |
| 9  | Elasticity Module (TPa)                    | ~1                   |
| 10 | Tensile strength                           | 130 GPa              |
| 11 | Young modulus                              | ~1 TPa               |

Atoms are one of the smallest building blocks of matter in the size range of about 1-3 nm. Atoms can come together to form nanoclusters. If at least one dimension of more than half of these structures that constitute a substance is smaller than 100 nm, the substance is called a nanomaterial (Binns, 2010). Nanocomposite comprises the combination of two or more different materials. At least one of these materials must be at the nanoscale (less than

100 nm). Nanocomposite materials have been found in many industries (Fesenko & Yatsenko, 2021; Li et al., 2021; Sen, 2020)

Powder metallurgy is an advanced material production method used for the processing of powdered metals. With this method, metal powders can be converted into cost-effective and specific products by passing through various processing stages. Complex structured parts that require high technology can be produced in sufficient quantity and high efficiency at the desired quality standards (German, 2016; Grande & Forno, 2016)

Electrical conductivity is a measure of a material's ability to conduct electricity. Properties such as structural defects of the material, temperature, and impurities can affect the conductivity. Generally, metals have high conductivity because there are more electrons that can move freely. Copper is a metal with very good electrical conductivity ( $6 \times 10^7 (\Omega \cdot m)^{-1}$  at room temperature). Therefore, it has become the basic component of the electrical-electronics industry today (Callister & Rethwisch, 2020)

In the International System of Units (SI), the conductivity is expressed in siemens per meter. However, in many studies, it is given as a percentage in the International Annealed Copper Standard (IACS). According to standard, the electrical conductivity of copper is accepted as 100%, and the electrical conductivities of other metals and alloys are calculated as percentages according to this value. Thus, a percentage expression proportional to the conductivity of Cu is obtained (Jones, 2013; Lu, 2004). There are some important basic issues to be considered in the production of metal-matrix composites. The strong structural properties of the additive materials and the formation of strong bonds with the metal components to be added can significantly contribute to the material development. However, graphene has a low interfacial affinity with copper. There is also a density difference between them. The homogeneous spread of the additive to the composite and the absence of agglomeration are important in terms of improving the mechanical properties of the composite. However, graphene tends to agglomerate when forming composites with Cu. Production methods and costs are also very important in composite design processes (Kumar & Xavior, 2014)

Most of the examined studies mainly aimed to improve the mechanical and electrical properties of copper. Although mechanical, thermal, or tribological properties have been improved in many studies (Akbarpour et al., 2020; Almonti et al., 2022; Dong et al., 2021; Fahimi & Abachi, 2021; Forati et al., 2021; Ghodrati & Ghomashchi, 2019; Guo et al., 2019; Han et al., 2020; 2021; Iqbal et al., 2020; Katarkar et al., 2021; Khamaj et al., 2021; Lasio et al., 2018; Pingale, Belgamwar & Rathore 2020a, 2020b; Yao et al., 2016; Zhang et al., 2019; Zhao et al., 2020) electrical properties have only been relatively improved (Chen et al., 2016; Huang et al., 2016; Jagannadham, 2012b; Varol et al., 2022; Xiong et al., 2015).

In this study, the electrical conductivities of composites formed from Cu and graphene were investigated with reference to consolidation methods and additive ratios. The results obtained from studies with the best conductivity values according to these two parameters are comparatively presented. Production methods are also mentioned. Thus, a preliminary idea was given to researchers interested in the subject before starting their studies.

## 2. Methodology

Table 2 shows the results of 14 different studies on copper/graphene composites according to production route, mixing method, and reinforcement rate.

**Table 2.** Results of studies on graphene reinforced copper composites.

| No | Route                        | Mixing type            | The reinforcement rate of best result (%) | Electrical conductivity (% IACS) | Rate of increase or decrease compared to the Pure Cu (%) | Reference                |
|----|------------------------------|------------------------|---|----------------------------------|--|--------------------------|
| 1  | Spark plasma sintering       | Sonication             | Cu/0.13 wt.% GNP–Ni                       | Composite: 92.9<br>Pure Cu: 99.1 | (- 6%)   | (Jiang, Zhou & Liu 2017) |
| 2  | Spark plasma sintering       | Stirring               | Cu 0.3 wt.% GNPs                          | Composite: 82.4<br>Pure Cu: 99.1 | (- 15%)  | (Jiang et al. 2016)      |
| 3  | Spark plasma sintering       | Molecular-level mixing | Cu 0.5 wt.% GNPs                          | Composite: 83.5<br>Pure Cu: 96.5 | (- 13.5%)  | (Si et al. 2017)         |
| 4  | Spark plasma sintering       | Wet mixing             | Cu-0.35 wt.% Gr                           | Composite: 90<br>Pure Cu: 98     | (- 8.16%)  | (Yang et al. 2022)       |
| 5  | Hot-pressing                 | Ball milling           | 1 wt. % GNS                               | Composite :94<br>Pure Cu: 77     | (+22%)   | (Salvo et al. 2019)      |
| 6  | High pressure torsion (HPT)  | Mechanical milling     | 10 wt. % Graphene                         | Composite: 87<br>Pure Cu: 98     | (- 11.22%)   | (Khobragade et al. 2019) |
| 7  | Hot isostatic pressing (HIP) | Wet mixing             | Cu-2 vol% GNPs                            | Composite: 77<br>Pure Cu: 78     | (- 1.28%)  | (Saboori et al. 2017)    |
| 8  | Spark plasma sintering       | Molecular-level mixing | Cu 0.2 vol% GNPs                          | Composite: 90<br>Pure Cu: 92.5   | (-3%)  | (F. Chen et al. 2016)    |
| 9  | Hot-pressing                 | Ball milling           | Cu 0.15 wt.% Ag-RGO                       | Composite: 93<br>Pure Cu: 81     | (+18%)   | (Luo et al. 2017)        |
| 10 | Hot-pressing                 | Preform impregnation   | Cu 1.2 vol% RGO                           | Composite: 98<br>Pure Cu: 96     | (+2%)  | (Xiong et al. 2015)      |
| 11 | Conventional sintering       | Ball milling           | Cu 0.5 wt.% GNPs                          | Composite: 78.6<br>Pure Cu: 93   | (- 15.5%)  | (Varol & Canakci 2015)   |
| 12 | Liquid-phase sintering       | Ball milling           | W70Cu30-0.5 wt.% GO                       | Composite: 45.7<br>Pure Cu: 42   | (+9%)  | (Akhtar et al. 2009)     |
| 13 | Electrodeposition            | Without stirring       | Cu-0.11 vol-fraction RGO                  | Composite: 97.7<br>Pure Cu: 81.8 | (+19%)   | (Jagannadham, 2012b)     |

|               |  |                             |                       |
|---------------|--|-----------------------------|-----------------------|
| 14            | Chemical vapor<br>deposition (CVD)                             | Composite: 98<br>(- 17.36%) | (Qiao et al.<br>2018) |
|               | +  |                             |                       |
|               | Catalytic<br>pyrolysis<br>(Pyrolysis<br>temperature<br>:800°C) |                             |                       |
|               | +  |                             |                       |
|               | Cold-rolled  |                             |                       |
|               | +  |                             |                       |
|               | Sintering  |                             |                       |
| Pure Cu: 83.5 |  |                             |                       |

The most important factors affecting the powder metallurgy process are the type of material, the mixing method used for the homogeneous distribution of the powder mixture, the method applied to produce the bulk material, and the sintering parameters. In addition, the conditions of the environment in which these steps take place should also be considered. For this reason, the methods used in some of the studies listed in Table 2 are mentioned.

According to Jiang et al. (2016), in previous studies, RGO (Reduced graphene oxide) was generally used as the graphene additive of copper-graphene composites. In this study, they showed that PG (pristine graphene) is a better graphene source than RGO. First, to prepare PG dispersion, PVP (polyvinyl pyrrolidone) was dissolved in water. To coat PG with PVP, a PG liquid suspension was added to the PVP solution and mixed for 30 min. Vacuum filtration was used to remove excess PVP from the water. An aqueous solution (3 wt.% PVA) was prepared from PVA and copper powders. Copper powder was added to the solution and mixed for 1 h. Vacuum filtration and rinsing were performed to remove excess PVA from the solution. Thus, PVA-adjusted copper powders were obtained. The resulting powders were divided into two groups in equal amounts. PVP-modified PG or GO solution was added dropwise to the aqueous solution produced with these powders and stirred. The stirring was stopped when the solution was colorless and transparent. The resulting product was rinsed after filtering. They were dried at 353 K for 12 h to obtain composite powders. Finally, the powders were processed at 573 K for 30 min and at 923 K for 2 h under gas flow; this mixture was a hydrogen-argon mixture, most of which was argon. A composite powder mixture was obtained. The samples were sintered by the spark plasma sintering method under 30 MPa pressure at 973 K for 5 min. Si et al. (2017) examined the effect of TiC (Titanium carbide) or VC (Vanadium carbide) coating on GNP–Cu composites. Some GNPs were mixed in ethanol for 1 h. The obtained liquid solution was then mixed with NaCl–KCl or LiCl–KCl molten by ball milling at 250 r/min speed for 12 hours. NaCl–KCl solution was used for the TiC coating of GNPs, and LiCl–KCl solution was used for the VC coating of GNPs. Drying and grinding processes were carried out to obtain a homogeneous mixture. After mixing this mixture with pure Cu powder, it was heated in an argon atmosphere in a quartz tube furnace at 850 °C for 1 h to obtain TiC-coated GNPs. To obtain the VC-coated GNPs under the same conditions, they were kept at 750 °C for 6 h. These products were mixed with Ti and GNPs at various concentrations after washing and drying. To obtain copper composite powders, GNP powders were mixed in an alcohol-containing solution containing copper ions. Then,  $C_6H_{12}O_6$  was added and mixed for 30 min. NaOH was slowly added to the solution, and stirring was continued. The solution was kept constant at 60 °C for 4 hours.

Processes such as washing and drying were applied to produce Cu-GNPs composites. Compressed composite powders were sintered by the spark plasma sintering method under a pressure of 35 MPa at 700 °C and a heating rate of 50 °C/min. Yang et al. (2022) mixed copper powder and wheat flour were mixed with ball-milling method. There is a 10:1 material ratio between the ball and powder. They were milled for 5 h at 423 rpm. Coated Cu powders were heated for 10 min in a vacuum tube furnace containing Ar (100 sccm) and H<sub>2</sub> (40 sccm) gases at 800 °C with a heating rate of 10 °C /min. Three different weight ratios of Gr-Cu composite powder were produced. The produced composite powders and pure Cu powder were then mixed by a wet mixing method. The samples were sonicated with ethanol at room temperature for 1 h. Powder solution stirred at 65 °C at 800 rpm. During mixing, ethanol was evaporated, and a homogeneous mixture was obtained. Finally, they were sintered by the spark plasma sintering method under 45 MPa pressure at 650 °C for 8 min. Saboori et al. (2017) used a wet mixing method for the homogeneous distribution of GNPs (Rashad et al. 2015). Copper and GNP powders were ultrasonicated in ethanol for 45 min. The GNP slurry was combined with the Cu powder suspension dropwise. After powder blend was ultrasonicated for 1 h, it was filtered and dried at 80 °C for 6 h. Composite powders were consolidated in dies to produce bulk samples. The samples were then sintered at 950 °C for 2.5 hours under N<sub>2</sub> atmosphere. Wang et al. (2016) obtained graphene, which was used in their studies, according to modified Hummers methods Ong et al. (2012). After processing the graphene, it was sonicated in an ethyl alcohol solution for several hours. Copper powder was added to the liquid suspension mixture and stirred for 20 h. The mixture was heated at 80 °C and dried under vacuum for 24 h. The consolidation process took place in two stages. First-stage composite powders were vacuum-sintered at 950 °C. In the second stage, the sample was hot pressed at 600 °C under 30 MPa pressure. Luo et al. (2014) used a one-step reduction method to produce Ag-RGO powder blend from GO (graphene oxide) and AgNO<sub>3</sub> (silver nitrate) Ji et al. (2015). Ag-RGO and Cu powders were mixed using the ball milling method which applied at 500 rpm rotation speed of 500 rpm for 5 h. Treated powders were sintered at 800 °C under pressures ranging from 30 to 60 MPa by the uniaxial hot-pressing method under vacuum conditions.

### 3. Results and Discussions

The results of the eight studies with the best electrical conductivity among the studies listed in Table 2 and the factors affecting the electrical conductivity are examined. In Table 3, these studies are presented together with data on production methods, mixing types, and reinforcement ratios.

**Table 3.** Best eight electrical conductivity results

| No | Route             | Mixing type      | The reinforcement rate of best result (%) | Electrical conductivity (% IACS) | Rate of increase or decrease compared to the Pure Cu (%) | Reference            |
|----|-------------------|------------------|---|----------------------------------|--|----------------------|
| 1  | Hot-pressing      | Ball milling     | 1 wt. % GNS                               | Composite :94                    | (+22%)   | (Salvo et al., 2019) |
| 2  | Electrodeposition | Without stirring | Cu-0.11 vol% RGO                          | Composite: 97.7                  | (+19%)   | (Jagannadham, 2012b) |
| 3  | Hot-pressing      | Ball milling     | Cu-0.15 wt. % Ag-RGO                      | Composite: 93                    | (+18%)   | (Luo et al., 2017)   |



|   |                              |                        |                     |                 |           |                        |
|---|------------------------------|------------------------|---------------------|-----------------|-----------|------------------------|
| 4 | Liquid-phase sintering       | Ball milling           | W70Cu30-0.5 wt.% GO | Composite: 45.7 | (+9%)     | (Akhtar et al., 2009)  |
| 5 | Hot-pressing                 | Preform impregnation   | Cu 1.2 vol% RGO     | Composite: 98   | (+2%)     | (Xiong et al., 2015)   |
| 6 | Hot isostatic pressing (HIP) | Wet mixing             | Cu-2 vol% GNPs      | Composite: 77   | (- 1.28%) | (Saboori et al., 2017) |
| 7 | Spark plasma sintering       | Molecular-level mixing | Cu 0.2 vol% GNPs    | Composite: 90   | (-3%)     | (Chen et al., 2016)    |
| 8 | Spark plasma sintering       | Sonication             | Cu 0.13 wt.% GNP–Ni | Composite: 92.9 | (- 6%)    | (Jiang et al., 2017)   |

According to the values given in Table 3, Salvo et al. (2019) achieved a 22% increase in the electrical conductivity value of the sample sintered by the hot-pressing method using 1 wt.% GNS (graphene nano sheets). In addition, they preferred the ball milling method. Similar to previous studies, they observed the agglomeration of few-layer graphene nanosheets. Agglomerations occurred particularly within grain boundaries. However, it was lower than those of other studies. In this study, it is emphasized how variables such as consolidation temperature, application pressure, and application time, which are used in other methods using the hot-pressing method, affect the electrical conductivity and hardness properties. While changing the consolidation temperature and application pressure from the application parameters, the time was kept constant. As a result, the electrical conductivity was observed to increase when the sintering temperature was slightly reduced and the application pressure was slightly increased. Jagannadham (2012a) obtained a 19% increase in the electrical conductivity value of the Cu-Gr composite films using the electrodeposition method. In addition, the authors used 0.11 vol% RGO (reduced graphite oxide). During the application of this method, magnetic stirring was used as the mixing method. When the mixing process was applied, the accumulation of graphene on the composite film increased and the electrical conductivity decreased. When magnetic stirring was not applied, the accumulated mass decreased and the electrical conductivity increased. Luo et al. (2017) observed a 18% increase in the electrical conductivity of the pattern sintered by the hot-pressing method when 0.15 wt.% Ag-RGO was used. In this study, silver was found to increase the interfacial bonding between graphene and copper. For the five different samples, the applied pressure was varied by keeping the temperature and time constant. Thus, the best electrical conductivity was obtained at an appropriate pressure (50 MPa). Akhtar et al. (2009) achieved a 9% increase in the electrical conductivity of a specimen sintered by liquid-phase sintering using 0.5 wt.% GO. Graphene was added to a pure W70Cu30 composite powder with an electrical conductivity of IACS 42%. The highest electrical conductivity value (~46% IACS) was obtained at a 0.5 wt.% graphene additive. After this value, the conductivity decreased rapidly. When the graphene additive content was 1 wt.%, WC and W<sub>2</sub>C (tungsten carbide) phases formed because of the high carbon content. The formation of these phases negatively affects the electrical conductivity. After the ball milling and sintering processes, some of the graphene was dispersed on the WCu composite powder and retained its form, but some of it was damaged by losing its structure. Xiong et al. (2015) achieved a 2% increase in the electrical conductivity of the sample sintered by the hot-pressing method using 1.2 vol% RGO. Inspired by the natural mother-of-pearl structure, the authors designed a skeleton similar to a brick-and-mortar structure. In this structure, rGrO was used as the brick, and Cu was used as the mortar. Copper was chosen for its enhanced electrical properties. Thus, both the mechanical properties and electrical conductivity were increased by the formation of the copper layer in the intermediate layer. Saborri (2017) obtained a 1.28% decrease in the electrical conductivity of the

Cu-GNPs composite films sintered by hot isostatic pressing (HIP) using 2 vol% GNPs (Graphene nanoplatelets). Using other studies, the author thinks that the decrease in the size of the grains or the large number of pores, dislocations, and voids in the composite may have made electron transfer difficult (Akhtar et al., 2009; Rajkumar & Aravindan, 2013). Two different reconsolidation processes were applied. As a result of these processes, a decrease in the number of pores and a slight increase in grain size were observed. The electrical conductivity did not substantially change until the addition of 2.0 vol of graphene, after which it began to decline. F. Chen et al. (2016) observed a 3% decrease in the electrical conductivity value of the pattern sintered by the spark plasma method when 0.2 vol% GNPs were used. In the present study, there was no significant decrease in electrical conductivity (85% of pure copper) until the graphene contribution was 0.4 vol.%. After this value, there was a significant decrease. The amount of graphene additive is important in this sense. The authors speculate that the reduced electrical conductivity is due to a decrease in grain size and an increase in dislocation density. In addition, the weak affinity between Cu and graphene and the gaps formed as a result of sintering increased the insulator (Nan et al., 1998; Yu et al., 2022). Jiang (2017) observed a 6% decrease in the electrical conductivity value of a specimen sintered by the spark plasma method using 0.13 wt.% GNP–Ni. In this study, untreated graphene was coated with Ni via electrolysis. Afterwards, these powders were mixed with pure Cu powders and used in the production process. The main purpose is not to experience much loss in electrical conductivity while increasing the mechanical properties. Previous studies have shown that oxygen has a negative effect on electrical conductivity. In this study, Ni is assumed to act like oxygen and has a negative effect on electrical conductivity.

#### 4. Conclusions

There are some important findings that stand out in studies in which the best conductivity results were obtained. The conclusions drawn according to the data obtained with reference to Table 3 are listed below.

- The most important factors affecting the electrical conductivity are graphene deposition at grain boundaries, oxide formation, dislocations, and pores. The selection of parameters for the production conditions and material properties that minimize the effects of these factors is important for the objectives of the studies.
- The best sintering methods in terms of the electrical conductivity of copper graphene composite materials are hot pressing and electrodeposition.
- When similar sintering and mixing methods are applied, if the material effect is not considered, when Ag is added to the graphene additive material, there is a significant decrease in the amount of graphene to be added.
- In Cu graphene composites, an increase in electrical conductivity was observed when Ag (silver) was added to the graphene.
- When hot pressing is chosen as the production method, the most effective mixing method is ball milling. However, other mixing methods can also be used.
- Although using oxide component materials such as RGO or GO as additive materials can have a negative effect on the electrical conductivity due to the oxygen that may arise, this problem can be overcome with composite production methods.



- When graphene and Ni (nickel) are used together as additive materials, a decrease in the electrical conductivity occurs.
- If the hot-pressing method is selected, the pressure applied will be very important.
- The presence of tungsten carbide (WC and W<sub>2</sub>C) in copper graphene composite materials reduces the electrical conductivity, but there is a serious decrease in the graphene-to-graphene ratio. This situation can be evaluated in terms of cost.
- To increase both mechanical and electrical properties, it may be appropriate to develop hybrid methods that include both or one of the hot-pressing and electrodeposition methods.
- In terms of graphene additive efficiency, spark plasma sintering and electrodeposition are among the best production methods.

In the future, detailed studies can be carried out with the properties of Cu-GNPs composites such as wear and corrosion. Conductivity changes can be studied by adding different reinforcement materials to these composites. In addition, the theoretical background of electrical conductivity can be added to pave the way for new studies. Studies can also be carried out on customized products considering the requirements of the industry. For instance, physical changes in switching elements can be studied in electrical devices where electrical conductivity is important.

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### Declaration of Competing Interest and Ethics

The authors declare no conflict of interest. This research study complies with research publishing ethics. The scientific and legal responsibility for manuscripts published in OPS Journal belongs to the authors.

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