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An improvement of current driving and electrical conductivity properties in covetics

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Abstract: Compared to pure Cu, Cu lattice retaining carbon atoms, called a covetic material, can have better electrical conductivity. Furthermore, the incorporation of carbon nanostructures into Cu-alloys could improve the mechanical properties of Cu-alloys. In the simulation study, we investigated Joule heating due to applied DC current on molten Cu metal concerning how to improve current density of covetic materials. In addition, we will discuss interfacial effects on covetic-metal electrodes to meet better current driving performance. The covetic composite excited at one electrode (width = 10 nm) has a higher current drive capability as a value of $3.54 \cdot 10^7$ A/m², for 1000 A current at a temperature of 1073.2 K, this value is a constant while temperature is changing up to 1573.2 K. We measured the conductivity of the proposed covetic materials at various carbon nanotube densities at room temperature. Experimental results show the lowest resistivity value accomplished after mixing and temperature annealing as a value of $1.78 \cdot 10^{-8}$ Ω.m, where the covetic sample has 1.27% carbon nanotube density, and that the electrical conductivity is superior to that of Cu-carbon nanotube composites previously reported.

Keywords: Covetics, current driving capacity, carbon nanotubes

1. Introduction

The production of a material with a higher conductivity at a lower density is inevitable in an application area of aviation and space flight systems [1]. Scherer [2] proposed the first "covetic" material form to increase the conductivity of Cu- and Al-based composites by mixing carbon in molten metal. Covalent bonding of carbon atoms into metallic matrix structure can be fabricated by using such a conventional induction furnace [3] or using pulsed laser deposition [4]. For instance, covetics fabrication using a conventional induction furnace, the molten Cu metal in graphite crucible is mixed by applying electrical current via one or more graphite electrodes, such as by grounding the outside of a crucible to form a DC current path.

In carbon incorporation mechanisms, activated carbon particles merged into a Cu metal matrix can provide better electric current driving performance, electrical and thermal conductivity, and material strength in comparison with such a base Cu metal [3, 5]. However, at this time of conducting this work, the process of merging carbon atoms into covetic composite has not been clarified. On

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the other hand, carbon incorporation into Cu lattice is insufficient due to low carbon solubility [6]. Some of carbon incorporation methods are electromigration and electro charge-assisted processes. In electromigration, ionized carbon atoms transform into a polymerized carbon structure. In electro charge-assisted process, carbon atoms bond to the molten Cu metal matrix via a high current through Cu-carbon nanotube (CNT) composite [7, 8].

In this work, we have studied the current driving capacity of covetics by performing simulation and experimental studies; as a result of increased Cu activation energy, covetics show high current density performance [9]. The current driving performance of covetics could be more effective up to 100 times [10] than its electrical conductivity performance. The simulation result of this study confirms limited performance of covetic conductivity compared to current density performance. The current driving performance of covetics has been also studied by considering thermal conductivity, CNT-electrode interface effect, and CNT breakdown mechanism. In the experimental study at room temperature, we measured the electrical conductivities of the proposed covetic material with different carbon nanotube densities.

2. Simulation setup

Physical and thermal simulations of the covetic material were performed in COMSOL Multiphysics. Finite element method (FEM) in COMSOL environment was applied to the thermal, electrical, and magnetic behavior of the proposed covetic material. The multiphysics simulation setup includes the following properties: joule heating, electric currents, electromagnetic heating, and heat transfer in solids. The activated features of "Electric currents" property in the simulation setup have features of current conservation, electric insulation, an input terminal, and an electric ground. A DC current is applied to a terminal and a ground placed on the left side of molten copper. It is worth saying simulations were realized in a nanoscale dimension, and a crucible was not modeled. The simulation parameters for the CNT material properties were obtained from [11–16], for the graphite material from [17–22], and for the molten copper from COMSOL default material library. The parameter values are given in Table 1.

Table 1: Material properties used in the simulations.

Property	Graphite	CNT
Density (kg/m^3)	1000	1500
Thermal conductivity ($W/(m.K)$)	704	3000
Heat capacity ($J/(kg.K)$)	650	700
Electrical conductivity (S/m)	6.10^5	2.10^7

The 2D geometric structure of the covetic devices that the different %areas of CNTs (circle shape, radius = 5 nm), the graphite electrode (width= 10 nm, height= 40 nm), and the molten copper (width = 50 nm, height = 40 nm, colored orange, are shown in Figures 1a and 1b.

2.1. The current driving capacity of covetic composite

The current driving capacity of the covetic material is calculated numerically via COMSOL to analyze the current driving capacity of the covetic material with the design parameters of CNTs, annealing temperature, applied DC current, and graphite electrode width and number.

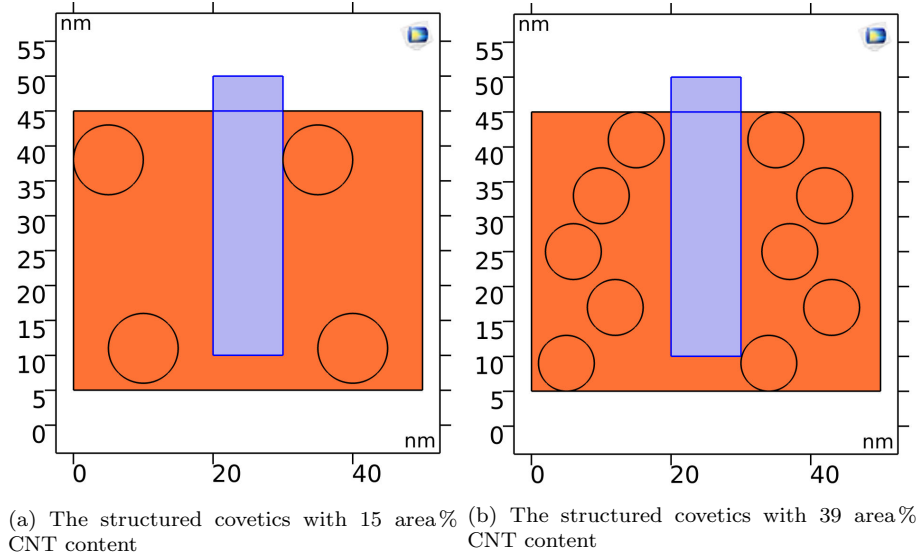


Figure 1: The 2D geometric structure of the proposed covetic material.

At 1073.2 K temperature, the current driving capacity of the covetic material with one electrode (width = 10 nm) and no CNT content is $0.24 \cdot 10^7 \text{ A/m}^2$ for 200 A current, and this value decreases to $0.22 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature. At 1073.2 K temperature, this value is $0.71 \cdot 10^7 \text{ A/m}^2$ for 600 A current, and at 1573.2 K temperature, this value is $0.67 \cdot 10^7 \text{ A/m}^2$. While it is $1.18 \cdot 10^7 \text{ A/m}^2$ for 1000 A current at 1073.2 K temperature, this value drops to $1.11 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature.

The current driving capacity of the covetic material, which includes one electrode (electrode width = 10 nm) and 15 area% CNT content (in Figure 2 as shown with acronym 4CNT), is $0.11 \cdot 10^7 \text{ A/m}^2$ for 200 A current at 1073.2 K temperature, and increases up to $0.13 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature. Based on a value of $0.32 \cdot 10^7 \text{ A/m}^2$ for 600 A current at 1073.2 K temperature, this value increases to $0.41 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature. While it is $0.54 \cdot 10^7 \text{ A/m}^2$ for 1000 A current at 1073.2 K temperature, this value increases to $0.67 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature.

In Figure 2, the covetic material setup, which includes one electrode (electrode width = 10 nm) and 39 area% CNT content (indicated by the acronym 10CNT), has current driving capacity as $0.71 \cdot 10^7 \text{ A/m}^2$ for 200 A current at 1073.2 K temperature, and this value remains unchanged up to $0.71 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature. At 1073.2 K temperature, the value for 600 A current is $2.12 \cdot 10^7 \text{ A/m}^2$, this value is constant up to 1573.2 K temperature. It is $3.54 \cdot 10^7 \text{ A/m}^2$ for 1000 A current at 1073.2 K temperature, and this value remains unchanged at higher temperature values up to 1573.2 K.

In order to advance current driving capacity, the covetic material with a 15 area% CNT content was examined in terms of different electrode widths and the number of graphite electrodes. For this evaluation, graphite electrode configurations are two electrodes (one of the electrode widths is 8 nm, acronym with2EW), two electrodes (one of the electrode widths is 4 nm, acronym with2EWby2), and one electrode (width = 10 nm, acronym withEWby2), as shown in Figure 3.

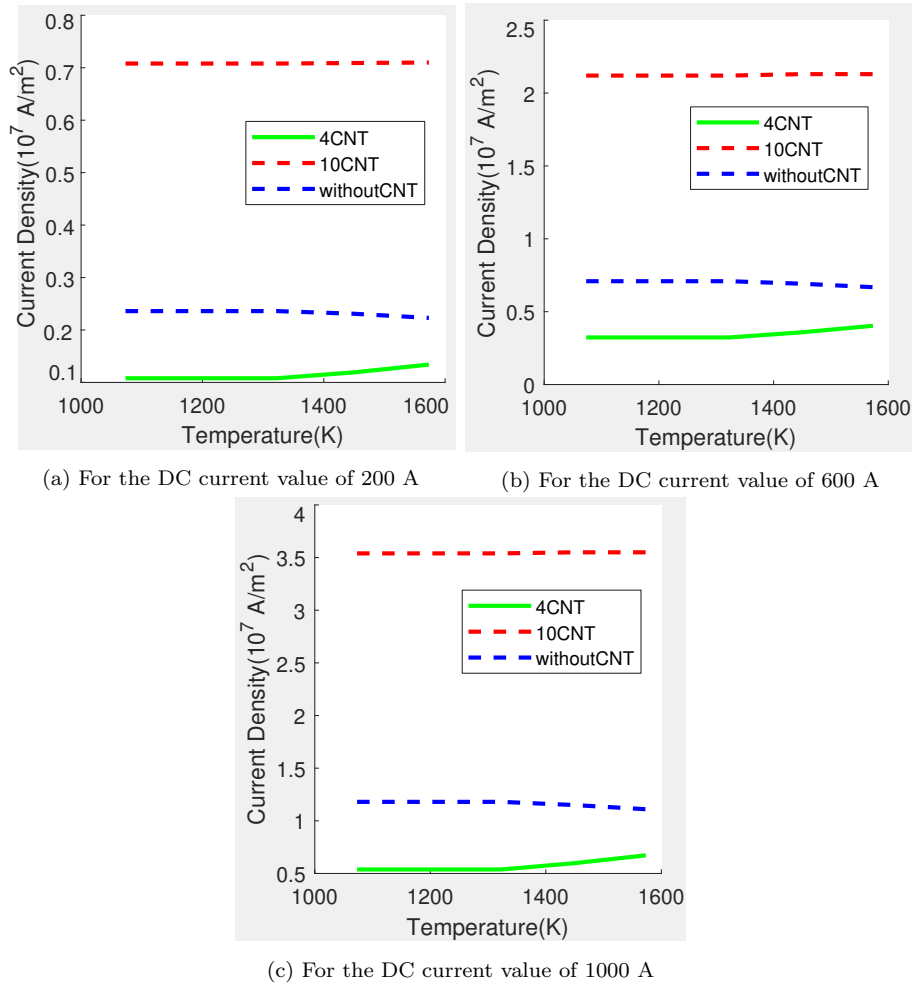


Figure 2: The current driving capacity values for various CNT contents in the covetic material.

At 1073.2 K temperature, the covetic material stimulated by the two electrodes (one of the electrodes is 8-nm wide) has a current driving capacity of $0.79 \cdot 10^7 \text{ A/m}^2$ for 200 A current, and at 1573.2 K temperature, the current driving capacity drops to $0.72 \cdot 10^7 \text{ A/m}^2$. The value for 600 A current is $2.38 \cdot 10^7 \text{ A/m}^2$ at 1073.2 K, but this value drops to $0.53 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K. While it is $3.97 \cdot 10^7 \text{ A/m}^2$ for 1000 A current at 1073.2 K temperature, this value drops to $3.65 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature.

The covetic material excited with the two electrodes (one of the electrode widths is 4 nm) has the current driving capacity as a value of $0.1 \cdot 10^7 \text{ A/m}^2$ for 200 A current at 1073.2 K temperature, and this value increases to $0.13 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature. A value of $0.32 \cdot 10^7 \text{ A/m}^2$ is obtained for 600 A current at 1073.2 K temperature. This value increases to $0.38 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature. While it is $0.54 \cdot 10^7 \text{ A/m}^2$ for 1000 A current at 1073.2 K temperature, this value increases to $0.63 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature.

The covetic material excited by a single electrode (electrode width is 4 nm) has a driving capacity for 200 A current of $0.13 \cdot 10^7 \text{ A/m}^2$ at 1073.2 K temperature, and a current driving capacity of 0.15

10^7 A/m^2 at 1573.2 K temperature. In the case of 600 A current, the value is $0.4 \cdot 10^7 \text{ A/m}^2$ at 1073.2 K temperature, and it increases to $0.45 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature. In the case of 1000 A current, the value is $0.66 \cdot 10^7 \text{ A/m}^2$ at 1073.2 K temperature, but it increases to $0.75 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K temperature.

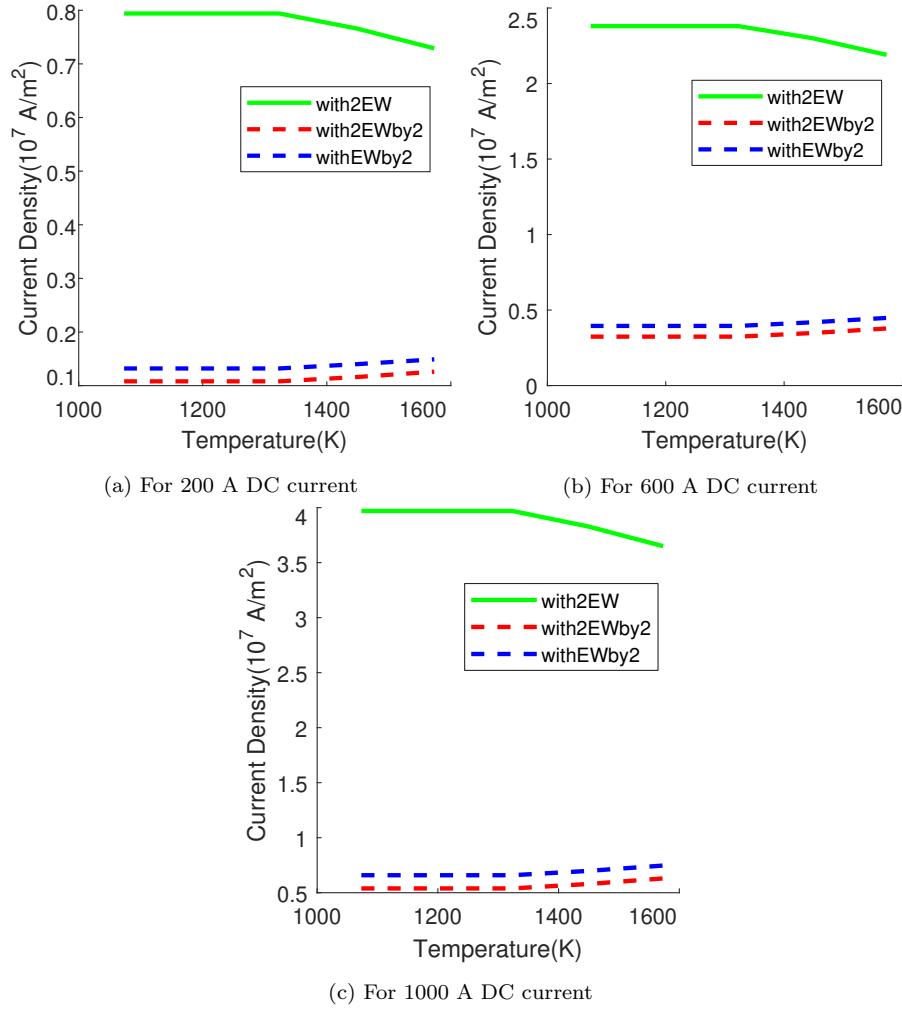


Figure 3: The current driving capacity values of the covetic material for different graphite electrode configurations, with 15 area% CNT content.

The covetic material, which has 39 area% CNT content, was examined in terms of different widths and the number of graphite electrodes for current driving capacity advancement. For this evaluation, graphite electrode configurations are two electrodes (one of the electrode widths is 8 nm, acronym with2EW), two electrodes (one of the electrode widths is 4 nm, acronym with2EWby2), and one electrode (width = 10nm, acronym withEWby2), as shown in Figure 4.

As a result of this evaluation, the covetic material excited with the two electrodes (one of the electrode widths is 8-nm) has a current driving capacity as a value of $2.32 \cdot 10^9 \text{ A/m}^2$ for 200 A current at 1073.2 K temperature, and this value slightly increases to $2.33 \cdot 10^9 \text{ A/m}^2$ at 1573.2 K temperature. In the case of 600 A current, the value is $6.95 \cdot 10^9 \text{ A/m}^2$ at 1073.2 K temperature, and it increases to

$7.0 \cdot 10^9 \text{ A/m}^2$ at 1573.2 K temperature. In the case of 1000 A current, the value is $11.6 \cdot 10^9 \text{ A/m}^2$ at 1073.2 K temperature, and it slightly increases to $11.7 \cdot 10^9 \text{ A/m}^2$ at 1573.2 K temperature.

The covetic material excited with the two electrodes (one of the electrode widths is 4 nm) has the current driving capacity of $1.58 \cdot 10^9 \text{ A/m}^2$ for 200 A current at 1073.2 K temperature, and this value remains constant up to 1573.2 K temperature. While it is $4.75 \cdot 10^9 \text{ A/m}^2$ for 600 A current at 1073.2 K temperature, this value drops to $4.71 \cdot 10^9 \text{ A/m}^2$ at 1573.2 K temperature. In the case of 1000 A current, it is $7.91 \cdot 10^9 \text{ A/m}^2$ at 1073.2 K temperature, but this value drops to $7.85 \cdot 10^9 \text{ A/m}^2$ at 1573.2 K temperature.

The covetic material excited with one electrode (the electrode width is 4 nm) has the current driving capacity as a value of $0.48 \cdot 10^7 \text{ A/m}^2$ for 200 A current at 1073.2 K temperature, and this value increases to $0.49 \cdot 10^7 \text{ A/m}^2$ value at 1573.2 K temperature. In the case of 600 A current, it is $1.46 \cdot 10^7 \text{ A/m}^2$ at 1073.2 K temperature, and this value remains unchanged up to 1573.2 K temperature. While it is $2.43 \cdot 10^7 \text{ A/m}^2$ for 1000 A current at 1073.2 K temperature, this value increases to $2.45 \cdot 10^7 \text{ A/m}^2$ at 1573.2 K.

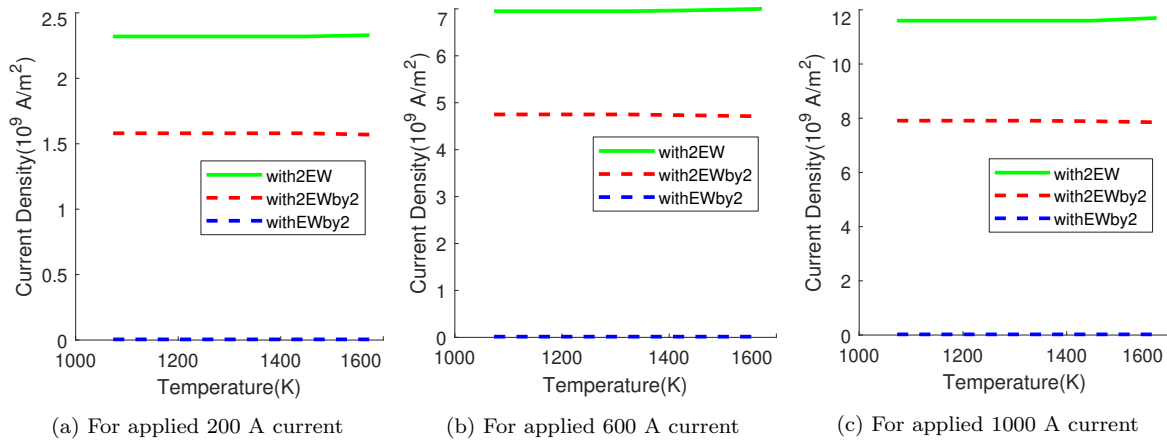


Figure 4: The current driving capacity values of the covetic material for different graphite electrode configurations, with 39 area% CNT content.

For the 15 area% CNT content and the 39 area% CNT content, the thermal conductivity parameter of CNTs have values from 2000 W/(m.K) to 6000 W/(m.K) t that shows us the current driving capacity of the covetic structure is not modulated with the thermal conductivity values.

2.2. The breakdown mechanism of the CNT composite

The CNT breakdown was studied in this work as a concern of breakdown effects on the current driving capacity of the covetic structure. Interface effects between CNTs and metal electrode [23, 24] are highly effective on material properties of which are contented CNTs such as the covetic composites considered here. The conductivity and current driving capacity can be enhanced by higher applied current values due to the joule heating effect [25]. The Joule heating causes contact resistance reduction between CNT and electrodes [26], which could be a possible reason for the current density improvement of the covetic composites. In another way, the current-carrying capacity of CNTs increases after breakdown due to reduced CNT resistivity [27] at applied higher DC stress. Increasing current flow (amperage)

into CNTs can decrease contact resistance between CNTs and electrodes [28].

The current driving capacity of the covetic composite considering 15 area% CNT content and 39 area% CNT content was calculated under Joule heating effects for DC current values of 200 A and 600 A. The current density of 15 area% CNT content in the covetic composite with various graphite electrode configurations is performed at 200 A current at 1073.2 K temperature and 600 A current at 1448.2 K temperature, as shown in Figure 5. The current density of 39 area% CNT content in the covetic composite with various graphite electrode configurations is performed at 200 A current and 1073.2 K temperature and 600 A current and 1448.2 K temperature, as shown in Figure 6. Figure 5 (15 area% CNT content) shows the results for the wider electrode widths (4-nm and 8-nm) for the applied 200 A DC current value: the CNT breakdown probability increases 44%. And for 600 A DC current value, the CNT breakdown probability increases 54%. Figure 6 (39 area% CNT content) shows the results for the wider electrode widths (4-nm and 8-nm) for the applied 200-A DC current value: the CNT breakdown probability increases 43%. And for 600 A DC current value, the CNT breakdown probability increases 46%. It is important to say that the analysis result of the breakdown simulation setup using a wider electrode width comparing the electrode width of 8 nm and 10 nm is inefficient, which includes applied 200 A and 600 A current values.

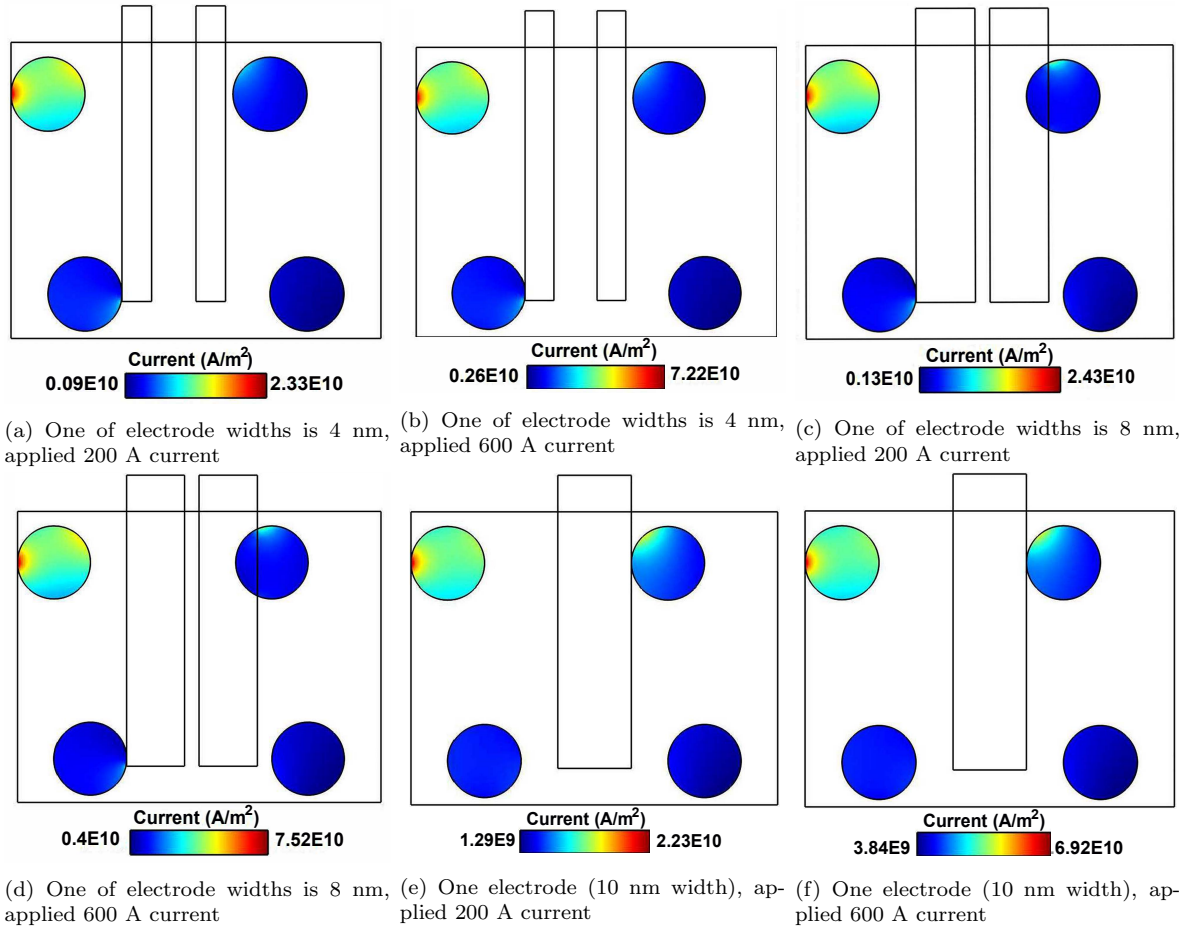


Figure 5: Breakdown current densities of 15 area% CNT content in the covetic composite.

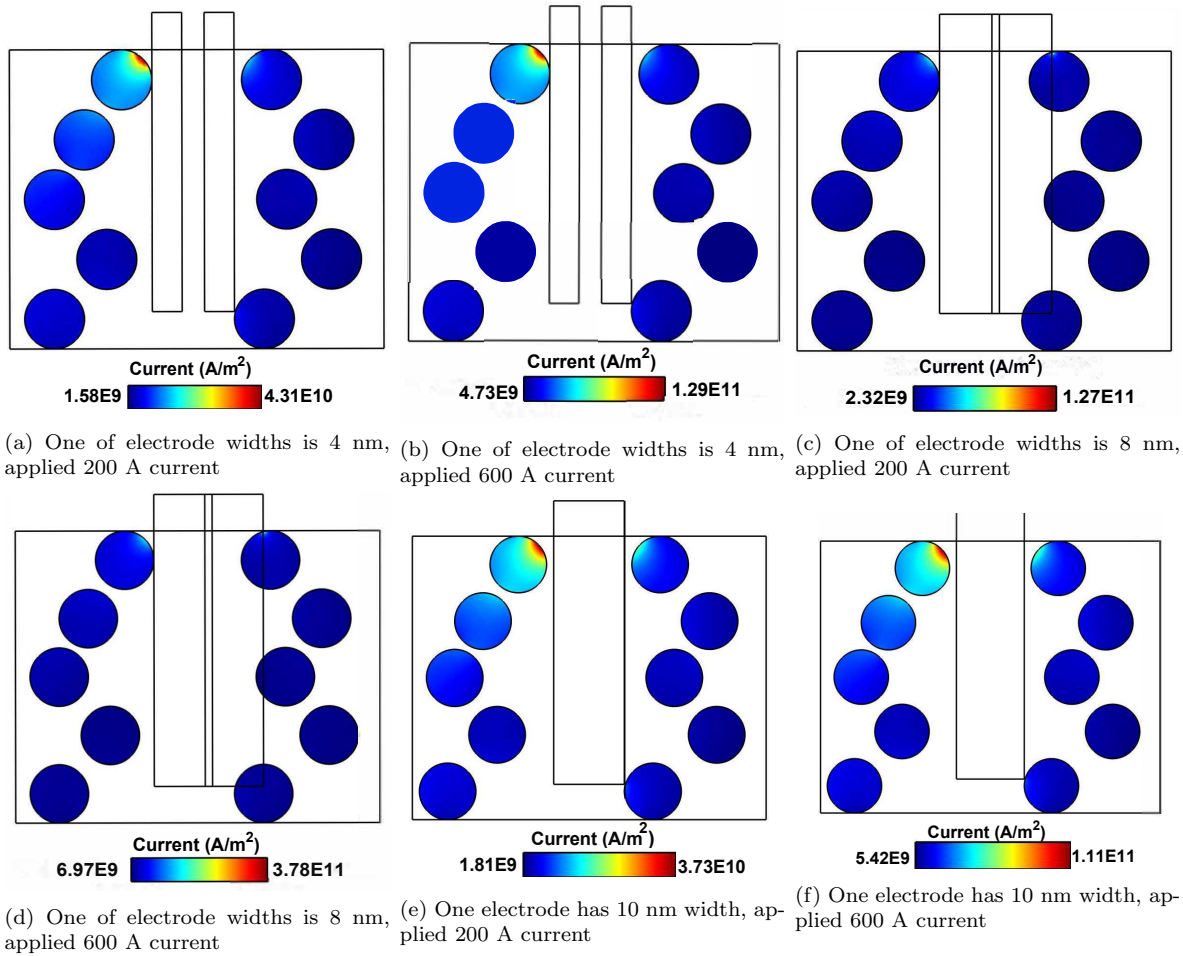


Figure 6: The breakdown current densities of 39 area% CNT content in the covetic composite.

The decrease of the breakdown power with increased electrode gap is expected for suspended CNTs [28]. The CNT breakdown current density is such an order of 10^8 A/cm^2 [29]. As a discussion of the breakdown simulation study, we examined CNT contents ranging from 15 to 39 area% in the covetic material allowing nearly two times higher breakdown current probability. Furthermore, we analyzed the CNT breakdown for the same CNT content for 200 A and 600 A applied DC current values that cause nearly two times higher breakdown current probability.

3. Experimental setup

Conductivity measurement tests of the covetic material with up to 6% variations were performed by probing different points of conductive cylinder rods. The four-point probe is a well-known method for conductivity measurement, the 4-point probe resistance equation can be written as follows:

$$\rho = \frac{I}{V} \frac{1}{2\pi S}$$

where I is excitation current, V is measured voltage, and S is the distance between probes. CNT and Cu content mixed mechanically before melting. It is beneficial to distribute CNTs homogeneously in

the melt structure. Homogeneous mixing of Cu-CNT composite can be more effective by using granular Cu. In different experiments to observe the effect of an applied DC current given the experimental setup in Figure 7 published partially in [30], the casting process was repeated for DC current values between 100 A and 400 A during 10 min period. Table 2 shows conductivity results of the covetic sample without mixing process under applied different current signals. As a result, the resistivity of the covetic sample increases as a comparison with pure Cu.



Figure 7: The experimental preparation of the Cu-covetic material.

Table 2: The experimental results of the covetic samples were prepared without mixing process.

Property	Resistivity ($10^{-8}\Omega.m$)
Pure Cu	2.482
Cu-CNT with applied 200 A	5.174
Cu-CNT with applied 300 A	4.544

The covetic sample was prepared by mixing 1.27% CNT content; the resistivity enhances up to 10.16 % in proportion to pure Cu resistivity, where the value of DC current was 300 A, given in Table 3.

Table 3: The experimental results of the covetic samples were prepared by mixing 1.27 % CNT content.

Property	Resistivity ($10^{-8}\Omega.m$)
Pure Copper	2.015
Cu-CNT with applied 200 A	9.822
Cu-CNT with applied 300 A	1.829

However, the mixing process for resistivity advance is insufficient when the covetic sample has a higher Cu density, such as a value of 4.2 % as given in Table 4. The lowest resistivity value is $1.77810^{-8} \Omega.m$, where the covetic sample has 1.27% CNT density, prepared with mixing process and annealing process.

Table 4: The experimental results of the covetic samples were prepared by mixing 4.2 % CNT content.

Property	Resistivity ($10^{-8}\Omega.m$)
Pure copper	2.015
Cu-CNT with applied 150 A	10.256
Cu-CNT with applied 300 A	6.264

4. Conclusion

In this paper, the low resistivity Cu-CNT covetic material was achieved via casting process. To study the electrical, thermal, and geometric properties of the covetic material, we developed a simulation model of the proposed covetic material in COMSOL. The current driving capacity of the covetic material is calculated numerically via COMSOL. On the current driving capacity of this material, we examined the effects of different densities of area% CNT, CNT breakdown mechanism, various widths, and the number of graphite electrodes. Moreover, the CNT breakdown was studied as a concern of breakdown effects on the current driving capacity of the covetic structure. The current driving capacity of the proposed covetic material is not modulated thermal conductivity values of the CNT as a result of the simulation results. The simulation study results show the covetic composite excited with one electrode (width = 10 nm) has a higher current driving capacity as a value of $3.54 \cdot 10^7 A/m^2$ for 1000 A current at 1073.2 K temperature, and this value is constant up to 1573.2 K temperature. This result is improved to a value of $7.91 \cdot 10^9 A/m^2$, where the covetic composite with the same 39 area% CNT content but was excited with two electrodes (one of electrode width is 4 nm) by driving 1000 A current at 1073.2 K temperature. In the experimental study, the lowest resistivity value is obtained as a value of $1.778 \cdot 10^{-8} \Omega.m$, where the covetic sample was prepared 1.27% CNT density with mixing process and with annealing process. As a result, interface effects on CNTs and metal electrodes can be a possible reason for the current density improvement of the covetic composites. The Cu-covetics with low-resistivity has potential application area in naval and aerospace industry.

5. Acknowledgment

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