

Influence of homogenization heat treatment on grain characteristics and mechanical properties of copper-silicon-zinc and copper-silicon-tin ternary alloys

Nkem Emelike Nwankwo¹, Kingsley Chidi Nnakwo², Agatha Ifeoma Ijomah^{3*},
Ifeanacho Uchenna Okeke⁴

Department of Metallurgical and Materials Engineering, Nnamdi Azikiwe University, Awka, Nigeria

Corresponding author address: ai.ijomah@unizik.edu.ng

DOI: <https://doi.org/10.5281/zenodo.8348507>

Published Date: 15-September-2023

Abstract: The main objective of this research is to explore the influence of homogenization heat treatment on grain characteristics and mechanical properties of copper-silicon-zinc and copper-silicon-tin ternary alloys. The alloys compositions were designed using response surface optimal design (RSOD). The designed alloy compositions were melted, cast, and subjected to homogenization at 900°C for 5 h. The grain characteristics of the produced alloys were analyzed using an optical microscope (OM). The properties investigated were percentage elongation, tensile strength, and hardness. The OM results revealed the presence of segregated primary silicon and coarse intermetallic phase in the parent alloy. The surface morphology of the doped alloy consisted of refined and modified intermetallic phases evenly dispersed in the alloy structure. The mechanical tests results showed that the percentage elongation, ultimate tensile strength and hardness of the alloy increased significantly by addition of tin and zinc. Homogenization heat treatment led to further increase in the ultimate tensile strength and hardness of Cu-3Si-3Zn alloys from 353 MPa to 376 MPa and from 254 BHN to 268 BHN. Cu-3Si-3Sn ternary alloys showed decreasing trends in ultimate tensile strength and hardness values after undergoing homogenization heat treatment. However, it recorded an increased percentage elongation after homogenization heat treatment.

Keywords: Cu-3Si-3Zn; Cu-3Si-3Sn; grainsmorphology; strength; hardness.

1. INTRODUCTION

Copper is a widely used metal, particularly in various industrial and electronic applications, due to its unique combination of properties. Copper is renowned for its high electrical conductivity, which makes it an ideal choice for electrical connectors, lead frames, and micro-electronic devices (Nnakwo, 2019; Nnakwo et al., 2017a,b; 2019a,b; 2020, 2021, 2022; Nnakwo and Nnuka, 2018; Garbacz-Klempka et al., 2018). This property allows for efficient transmission of electrical signals. Copper is relatively low in cost compared to many other metals with similar electrical conductivity characteristics, making it a cost-effective choice for various applications. Copper possesses excellent ductility and malleability, making it suitable for applications where it needs to be shaped, bent, or formed into various components such as bolts, nuts, valves, and fittings (Qing et al., 2011; Xie et al., 2003; Lei et al., 2017; Gholami et al., 2017; Qian et al., 2017; Suzuki et al., 2006). Copper is often alloyed with other elements like silicon, tungsten, zinc, tin, magnesium, manganese, and nickel to enhance its properties. These alloying elements can increase strength and hardness while minimizing the reduction in electrical conductivity. Silicon, when added to copper, can improve its fluidity and hardness. However, it comes at the expense of reduced ductility and electrical conductivity. The addition of silicon induces the precipitation of hard but brittle phases, such as Cu₃Si, Cu₁₅Si₄, and Cu₅Si, when the material cools slowly to ambient temperature (Wang et al., 2016; Li et al., 2017; Pan et al., 2007; Li et al., 2009; Lei et al., 2013a, 2013c; Eungyeong et al., 2011; Ho et al., 2000).

Copper-silicon alloys are used as electrodes in lithium-ion batteries (Ketut et al., 2011). The addition of silicon can enhance the performance of these electrodes in terms of capacity and cycling stability. Copper-silicon alloys can also serve as catalysts in various chemical processes, such as the production of nanosized and nanotube zinc oxide rods (Pak et al., 2016; Mattern et al., 2007). They are also employed in the fabrication of musical equipment due to their excellent damping properties. These alloys help reduce vibrations and unwanted noise, making them suitable for musical instruments (Cai et al., 2011). Effects of various alloying elements on the enhancement of properties have been explored by various researchers. Nickel is one of the key alloying elements known to enhance the hardness and electrical conductivity of Cu-Si alloys (Qian et al., 2017; Suzuki et al., 2006; Wang et al., 2016; Pan et al., 2007; Li et al., 2009; Lei et al., 2013b; Eungyeong et al., 2011; Ho et al., 2000). Other elements like aluminium, chromium, iron, magnesium, and tin have also been used to modify Cu-Si alloys. For example, iron has been found to enhance both hardness and electrical conductivity, while chromium and zirconium induce microstructural refinement and the precipitation of specific intermetallic phases, leading to improved strength. Combined Addition of Chromium and Zirconium: Combining chromium and zirconium in nickel-doped Cu-Si alloys has been shown to result in alloys with excellent hardness and electrical conductivity. The strengthening of copper alloys is achieved through the precipitation of various phases, including β -Ni₃Si, α -Cu(Ni, Si), γ' -Ni₃Al, β -Ni₃Si, and δ -Ni₂Si. These phases form as a result of the alloying elements and subsequent aging process (Suzuki et al., 2006; Wang et al., 2016, 2018; Li et al., 2017; Wang et al., 2018).

The main objective of this research is to develop Cu-Si base alloys with improved ductility, tensile strength, and hardness through additions of zinc and tin; and subsequent homogenization heat treatment. This enhancement of properties aims to expand the potential applications of these alloys in various engineering sectors.

2. EXPERIMENTAL PROCEDURE

The Cu-3Si-3Zn and Cu-3Si-3Sn ternary alloys were prepared using analytical grades copper rods (99.8% pure), silicon powder (99.7% pure), zinc powder (98.5% pure), and tin powder (98.7% pure). The weight in gram of each material was determined, measured using an electronic compact scale (Model: BL20001), and charged into the platinum crucible pot in an inert gas atmosphere. The melt was cast into a steel mold of dimensions 250 x 16 mm² and cooled inside the steel mold to ambient temperature. The developed alloys were subjected to homogenization heat treatment at 900°C for 5 h using a tube furnace (TSH12/25024166CG) equipped with an external thermocouple ($\pm 1^\circ\text{C}$ accuracy). The tensile test samples were machined to the required dimensions with total length 120 mm, 50 mm gauge length, and 10 mm diameter. The samples surfaces were ground and polished adequately for hardness test using an electric grinder and aluminium oxide powder. The tensile strength test was determined using 100kN capacity automated tensile strength tester (Model: 130812) and the hardness measured using a Vickers hardness tester (Model: VM-50). The Vickers hardness was conducted at a load of 183.9kgf and dwelling time of 5s. The diagonals of indentations were measured using a 20X optical microscope (Olympus BH) and the mean diameter determined.

3. RESULTS AND DISCUSSION

Figs. 1-3 show the effects of homogenization heat treatment on the percentage elongation, ultimate tensile strength, and hardness of Cu-3Si-3Zn and Cu-3Si-3Sn ternary alloys. Fig. 1 showed that addition of zinc and tin to Cu-3Si alloy led to a significant increase in the percentage elongation. Zinc increased the percentage elongation from 9.4% to 17.4%, while tin recorded a remarkable 85.11% increase in percentage elongation of Cu-3Si alloy. This implies the resistance of these alloys to deformation and fracture. The increase in percentage elongation can be attributed to the solid solution of zinc and tin in the copper matrix. Both the parent alloy (Cu-3Si) and Cu-3Si-3Zn alloys experienced a decline in percentage elongation after homogenization heat treatment. In contrast, Cu-3Si-3Sn alloy exhibited an increased percentage elongation after heat treatment. Homogenization heat treatment had varying effects on ultimate tensile strength and hardness depending on the alloy composition (Figs. 2 and 3). Both Cu-3Si-3Zn and Cu-3Si alloys displayed higher ultimate tensile strength and hardness in the heat-treated conditions compared to their as-cast states. Cu-3Si-3Zn alloy's ultimate tensile strength increased from 353 MPa to 376 MPa, and its hardness increased from 254 BHN to 268 BHN after heat treatment. Cu-3Si alloy, on the other hand, showed a negative response to heat treatment, with a decrease in ultimate tensile strength from 238 MPa to 229 MPa and a decrease in hardness from 278 BHN to 251 BHN. The decline in mechanical properties of Cu-3Si alloy after heat treatment may be attributed to the precipitation of coarse grains in the copper matrix as shown in Fig. 4d.

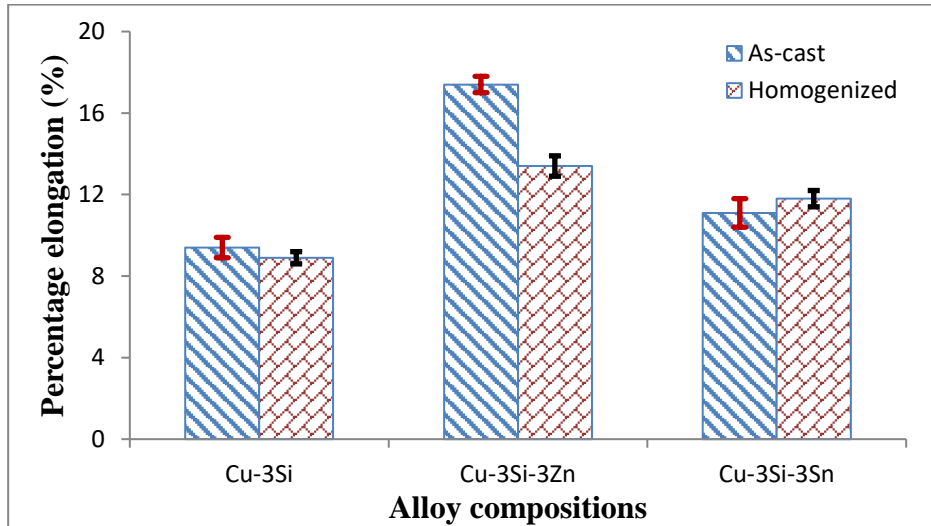


Fig. 1: Effect of homogenization on the percentage elongation of Cu-3Si-3Zn and Cu-3Si-3Sn ternary alloys

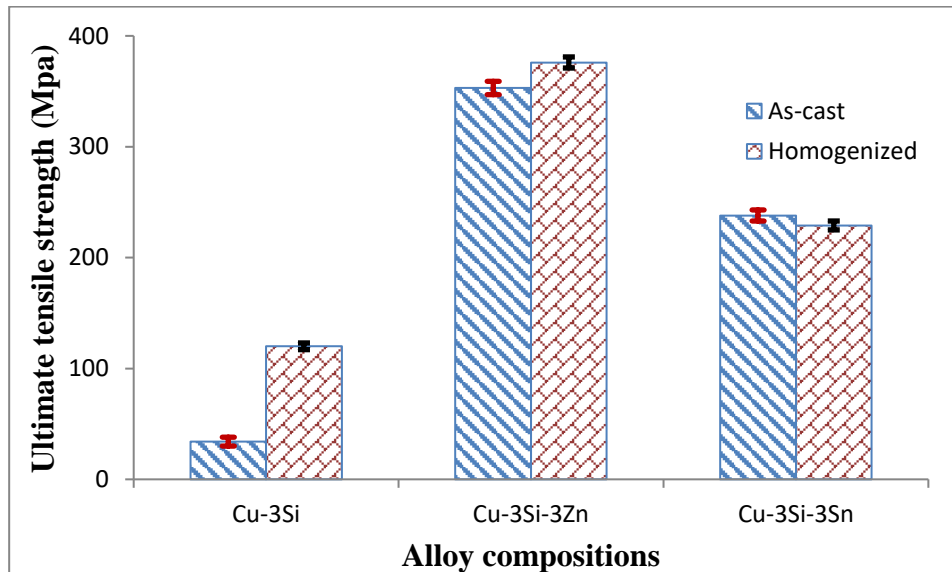


Fig. 2: Effect of homogenization on the ultimate tensile strength of Cu-3Si-3Zn and Cu-3Si-3Sn ternary alloys

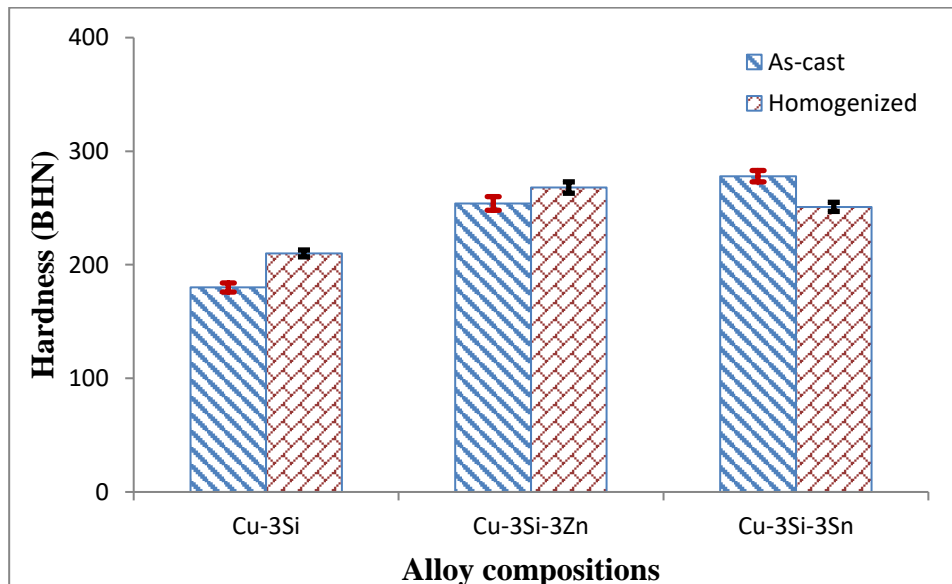


Fig. 3: Effect of homogenization on the hardness of Cu-3Si-3Zn and Cu-3Si-3Sn ternary alloys

Fig. 4 shows the optical microstructures of Cu-3Si-3Zn and Cu-3Si-3Sn ternary alloys in as-cast and homogenized conditions. Analysis of Fig. 4a shows grains of different morphologies separated by grain boundaries. These grains disappeared on the application of homogenization heat treatment as shown in the OM image of the Cu-3Si-3Zn ternary alloy (homogenized) (Fig. 4b). This indicates adequate solid solution of zinc in the copper matrix, showcasing the presence of α -phase. This can be linked with the increased ultimate tensile strength and hardness of the alloy as presented in Figs. 2 and 3. Comparative analysis of the surface morphologies of Cu-3Si-3Sn ternary alloys in as-cast and homogenized states show clearly the presence of plate-like coarse grains in the homogenized sample, indicating lower dislocation density in the alloy structure. This probably led to the observed decrease in the mechanical properties of the alloy compared to the as-cast sample.

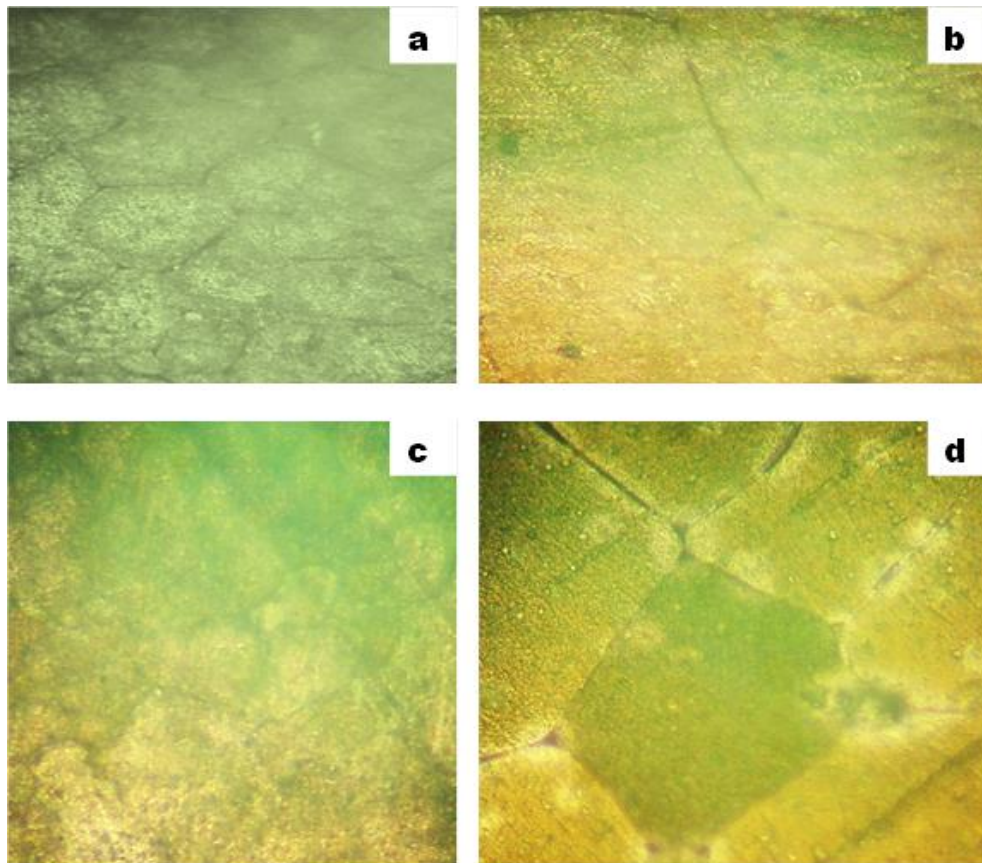


Fig. 4: Optical microstructure of (a) Cu-3Si-3Zn (as-cast)(b) Cu-3Si-3Zn (homogenized)(c) Cu-3Si-3Sn (as-cast)(D) Cu-3Si-3Sn (homogenized).

4. CONCLUSIONS

The influence of homogenization heat treatment on grain characteristics and mechanical properties of copper-silicon-zinc and copper-silicon-tin ternary alloys has been studied experimentally. The effects of zinc and tin additions on the microstructure, ultimate tensile strength, percentage elongation, and hardness of Cu-3Si alloys were also investigated. Results of the study demonstrated that the addition of zinc and tin to Cu-3Si alloy significantly improved its ductility, with tin being particularly effective in this regard. However, the response to homogenization heat treatment varies among the alloy compositions, with Cu-3Si and Cu-3Si-3Zn showing improvements in mechanical properties and Cu-3Si-3Sn experiencing declines due to grain coarsening. These findings have important implications for the development and application of these ternary copper alloys in engineering and manufacturing contexts, where a balance between strength and ductility is essential. Further research may be needed to optimize the heat treatment processes for specific alloy compositions and applications.

ACKNOWLEDGEMENT

The authors acknowledge the support of the management of Notex Electronics Nigeria Ltd and the management of Cutix Cable Plc, Nnewi Nigeria for providing equipment used for this research.

REFERENCES

- [1] Cai, H., Tong, D., Wang, Y., Song, X., Ding, B., 2011. Reactive synthesis of porous Cu₃Si compound. *J. Alloys Comp.* 509, 1672–1676. DOI: 10.1016/j.jallcom.2010.09.116.
- [2] Eungyeong, L., Seungzeon, H., Kwangjun, E., Sunghwan, L., Sangshik, K., 2011. Effect of Ti addition on tensile properties of Cu-Ni-Si alloys. *Met. Mater. Int.* 17 (4), 569–576. DOI: 10.1007/s12540-011-0807-7.
- [3] Garbacz-Klempka, A., Kozana, J., Piękoś, M., Papaj, M., Papaj, P., Perek-Nowak, M., 2018. Influence of modification in centrifugal casting on microstructure and mechanical properties of silicon bronzes. *Archives of Foundry Engineering.* 18, 11-18. DOI: 10.24425/123594.
- [4] Gholami, M., Vasely, J., Altenberger, I., Kuhn, H.A., Wollmann, M., Janecek, M., Wagner, L., 2017. Effect of microstructure on mechanical properties of CuNiSi alloys. *J. Alloy. Compd.* 696, 201–212. DOI: 10.1016/j.jallcom.2016.11.233.
- [5] Hines, W.W., Montgomery, D.C., Goldsman, D.M., Borror, C.M., 2003. *Probability and Statistics in Engineering*, 4th Edition, John Wiley & Sons, 2003.
- [6] Ho, J.R., Hyung, K.B., Soon, H.H., 2000. Effect of thermo-mechanical treatments on microstructure and properties of Cu-base lead frame alloy. *J. Mater. Sci.* 35 (14), 3641–3646. DOI: 10.1023/A:1004830000742.
- [7] Jung, S.J., O’Kelly, C.J., Boland, J.J., 2015. Position controlled growth of single crystal Cu₃Si nanostructures. *Cryst. Growth Des.* 15, 5355-5359. DOI: 10.1021/acs.cgd.5b00947.
- [8] Ketut, G.S.I., Soekrisno, R., Suyitno, M.I. Made, 2011. Mechanical and damping properties of silicon bronze alloys for music applications. *Int. J. Eng. Tech. IJETIJENS.* 11 (06), 81–85.
- [9] Lei, Q., Li, Z., Dai, C., Wang, J., Chen, X., Xie, J.M., Yang, W.W., Chen, D.L., 2013a. Effect of aluminium on microstructure and property of Cu–Ni–Si alloys. *Mater. Sci. Eng., A* 572, 65–74. DOI: 10.1016/j.msea.2013.02.024.
- [10] Lei, Q., Li, Z., Xiao, T., Pang, Y., Xiang, Q.Z., Qiu, W.T., Xiao, Z., 2013b. A new ultrahigh strength Cu-Ni-Si alloy. *Intermetallics* 42, 77–84. DOI: 10.1016/j.intermet.2013.05.013.
- [11] Lei, Q., Xiao, Z., Hu, W., Derby, B., Li, Z., 2017. Phase transformation behaviors and properties of a high strength Cu-Ni-Si alloy. *Mater. Sci. Eng., A* 697, 37–47. DOI: 10.1016/j.msea.2017.05.001.
- [12] Li, Z., Pan, Z.Y., Zhao, Y.Y., Xiao, Z., Wang, M.P., 2009. Microstructure and properties of high-conductivity, super-high-strength Cu-8.0Ni-1.8Si-0.6Sn-0.15Mg alloy. *J. Mater. Res.* 24 (6), 2123–2129. DOI: 10.1557/jmr.2009.0251.
- [13] Li, D., Wang, Q., Jiang, B., Li, X., Zhou, W., Dong, C., Wang, H., Chen, Q., 2017. Minor-alloyed Cu-Ni-Si alloys with high hardness and electric conductivity designed by a cluster formula approach. *Progress in Nat. Sci.: Mater. Int.* 27 (4), 467–473. DOI: 10.1016/j.pnsc.2017.06.006.
- [14] Mattern, N., Seyrich, R., Wilde, L., Baetz, C., Knapp, M., Acker, J., 2007. Phase formation of rapidly quenched Cu–Si alloys. *J. Alloy. Compd.* 429, 211–215. DOI: 10.1016/j.jallcom.2006.04.046.
- [15] Moon, T., Kim, Ch., Park, B., 2006. Electrochemical performance of amorphous-silicon thin films for lithium rechargeable batteries. *J. Power Sources.* 155, 391-394. DOI: 10.1016/j.jpowsour.2005.05.012.
- [16] Nnakwo, K. C., Okeke, I.U., Nnuka, E.E., 2017a. Structural modification and mechanical properties of Cu-3wt%Si-xwt%Sn alloy. *International Journal of Scientific Research in Science, Engineering and Technology.* 3, 184-187.
- [17] Nnakwo, K. C., Okeke, I.U., Nnuka, E.E., 2017b. Effect of zinc content on the structure and mechanical properties of silicon bronze. *International Journal of Scientific Research in Science, Engineering and Technology.* 3, 179-183.
- [18] Nnakwo, K. C., Nnuka, E. E., 2018. Correlation of the structure, mechanical and physical properties of Cu3wt%Si-xwt%Sn silicon bronze. *Journal of Engineering and Applied Sciences.* 13, 83-91.
- [19] **Nnakwo K. C.**, 2019. Effect of tungsten content on the structure, physical and mechanical properties of silicon bronze (Cu-3wt%Si), *Journal of King Saud University - Science*, 31(4), 844-848. doi: <https://doi.org/10.1016/j.jksus.2017.12.002>.

- [20] Nnakwo, K. C., Mbah, C. N., and Daniel-Mkpume, C. C., 2019a. Investigation of the structural sensitive behavior of Cu-3Si-xMn ternary Alloys. *Journal of King Saud University –Science*, 31(4), 1056-1063. <https://doi.org/10.1016/j.jksus.2019.01.001>.
- [21] Nnakwo, K. C., Mbah, C. N., and Nnuka, E. E., 2019b. Influence of trace additions of titanium on grain characteristics, conductivity and mechanical properties of copper-silicon-titanium alloys. *Heliyon*.5(10), e02471. <https://doi.org/10.1016/j.heliyon.2019.e02471>.
- [22] **Nnakwo**, K. C., Mbah, C. N., and Ude, S. N., 2020. Influence of chemical composition on the conductivity and on some mechanical properties of Mg-doped Cu-Si alloy. *Journal of King Saud University–Engineering Science*.32(5),287-292 <https://doi.org/10.1016/j.jksues.2019.03.005>.
- [23] **Nnakwo, K. C.**, Osakwe, F. O., Ugwuanyi, B. C., Oghenekowho, P. A., Okeke, I. U., & Maduka, E. A., 2021. Grain characteristics, electrical conductivity, and hardness of Zn-doped Cu–3Si alloys system. *SN Applied Sciences*, 3(11). <https://doi.org/10.1007/s42452-021-04784-1>.
- [24] **Nnakwo**, K. C., Odo, J. U., Eweka, K. O., Okafor, J. S., Ijomah, A. I., Maduka, E. A., and Ugwuanyi, B. C., 2022. Evaluation of the Electrical Conductivity and Mechanical Properties of Cu–3Ti–1.5Ni–0.5Si Quaternary Alloy, *JOM: the journal of the minerals, metals, and materials society*, Vol. 74, (Issue 5); 4174-4180.
- [25] Pak, A.Y., Shatrova, K.N., Aktaev, N.E., Ivashutenko, A.S., 2016. Preparation of ultrafine Cu₃Si in high-current pulsed arc discharge. *Nanotechnol. Russ.* 11 (9– 10), 548–552. DOI: 10.1134/S199507801605013X.
- [26] Pan, Z.Y., Wang, M.P., Li, Z., 2007. Effect of trace elements on properties of Cu-Ni-Si alloy. *Mater. Rev.* 21 (5), 86–89.
- [27] Polat, B.D., Eryilmaz, O.L., Keleş, O., Erdemir, A., Amine, K., 2015. Compositionally graded SiCu thin film anode by magnetron sputtering for lithium ion battery. *Thin Solid Films*.596, 190–197. DOI: 10.1016/j.tsf.2015.09.085.
- [28] Qian, L., Zhou, L., Zhou, L., Yang, G., Xi, P., Benjamin, D., 2017. Microstructure and mechanical properties of a high strength Cu-Ni-Si alloy treated by combined aging processes. *J. Alloy. Compd.* 695, 2413–2423. DOI: 10.1016/j.jallcom.2016.11.137.
- [29] Qing, L., Li, Z., Wang, M.P., 2011. Phase transformation behavior in Cu–8.0Ni–1.8Si alloy. *J Alloy Compound*. 509 (8), 361-367. DOI: 10.1016/j.jallcom.2010.12.115.
- [30] Suzuki, S., Shibusani, N., Mimura, K., Isshiki, M., Waseda, Y., 2006. Improvement in strength and electrical conductivity of Cu–Ni–Si alloys by aging and cold rolling. *J. Alloy. Compd.* 417 (1–2), 116–120. DOI: 10.1016/j.jallcom.2005.09.037.
- [31] Wang, W., Kang, H., Chen, Z., Chen, Z., Li, R., Yin, G., Wang, Y., 2016. Effects of Cr and Zr addition on microstructure and properties of Cu-Ni-Si alloys. *Mater. Sci. Eng., A* 673, 378–390. DOI: 10.1015/j.msea.2016.07.021.
- [32] Wang, W., Guo, E., Chen, Z., Kang, H., Chen, Z., Zou, C., Lia, R., Yina, G., Wang, T., 2018. Correlation between microstructures and mechanical properties of cryorolled CuNiSi alloys with Cr and Zr alloying. *Materials Characterization*. 144, 532–546. DOI: 10.1016/j.matchar.2018.08.003.
- [33] Xie, S.S., Li, Y.L., Zhu, L., 2003. Progress of study on lead frame copper alloy and its implementation in electronic industry. *Rare Metals*.27, 76-79.
- [34] Xu, K., He, Y., Ben, L., Li, H., Huang, H., 2015. Enhanced electrochemical performance of Si-Cu-Ti thin films by surface covered with Cu₃Si nanowires. *J. Power Sources*. 281, 455-460. DOI: 10.1016/j.jpowsour.2015.02.023.