

STUDY ON THE INFLUENCE OF CONTINUOUS CASTING PARAMETERS ON QUALITY OF COMPOSITES Cu-C FOR THE POSSIBILITY OF THEIR PROCESSING IN DRAWING PROCESS

GNIEŁCZYK Marek, KNYCH Tadeusz, MAMALA Andrzej, SMYRAK Beata, KWAŚNIEWSKI Paweł, KIESIEWICZ Grzegorz, KAWECKI Artur, JABŁOŃSKI Michał, KORZEŃ Kinga, JURKIEWICZ Bartosz, FRAN CZAK Krystian, KOWAL Radosław, ZASADZIŃSKA Małgorzata

*AGH University of Science and Technology, Faculty of Non-Ferrous Metals,
Department of Metal Working and Physical Metallurgy of Non-Ferrous Metals,
Cracow, Poland, EU
Mgnielcz@agh.edu.pl*

Abstract

Observed in recent years - trends in the search for novel materials with unconventional properties arise from the constantly growing needs of the economy. An example of such a material is graphene, which in recent years has revolutionized the market and created opportunities to develop new composites based on graphene. Work on the creation of composite copper - graphene have been taken to improve the - first of all - the electrical conductivity of materials, but also strength and other performance properties. Are now being made various attempts to synthesize graphene and copper, among others, using electro-deposition processes, casting processes and the processes of consolidation of powders. It is suspected that the increase in electrical conductivity can be obtained in the case of the structure that is targeted. One in which graphene layers are arranged in the direction of conduction of electric current. Certainly, to obtain such a structure facilitates the process of continuous casting, and further processing the rods in the process of drawing further directs structure.

Keywords: Copper-graphene composites, electro-deposition process, casting

1. INTRODUCTION

Following common trends, researchers around the world are working on the invention of a unique material, characterized by technological superiority over existing materials. Due to the fact that the design of overhead lines commonly uses copper cables, efforts have been made to create composites based on that, armoring the base material with carbonaceous materials such as graphene [1-4], which thanks to its unique properties [5-6], give a great chance to create a material that combines their major features into a Cu-C composite in metallurgical synthesis. An advantage of graphene, which makes it stand out among other allotropic forms of carbon, is that it is not prone to form agglomerates (as nanotubes are) and has much greater active blending area with the matrix than nanotubes. Moreover, global trends related to application of carbon bring newer and cheaper technologies of acquiring this type of raw material. To that end, leading research and implementation centres works on development of methods of manufacturing and processing of aluminium-graphene or copper-graphene composites shown in **Table 1**. Such works are conducted in the following centres: US Army Benét Laboratories, Armaments Research Development and Engineering Center, Watervliet, Rice University and Rensselaer Polytechnic Institute in USA. Nowadays the only application of graphene prepared for common use in the near future is in touch screens developed by the Korean Samsung [1-6].

Table 1 Review of metal-graphene synthesis methods [1-6]

No.	Synthesis method	Research centres	Estimated/achieved conductivity, % IACS (International Annealed Copper Standard)
1	Electro-deposition on the Cu surface	NanoRidge USA, AGH WMN	130
		University of Central Florida	200
2	Extrusion and die-casting of powders	Cleveland State University	1000
3	Ultrasonic coating with carbon materials	Los Alamos Nat'l Lab	100
4	Composites made of CNT and Cu fibres	Cambridge University, AGH WMN	Not known
5	Metallurgy of Cu-C composites	Third Millennium Materials, AGH WMN	102
6	CVD laser deposition	Los Alamos Nat'l Lab	10 000
7	Metallurgy of powders	Sumitomo, Siemens, Plansee, Fraunhofer, Hokkaido U., KAIST	100

Synthesis of graphene with aluminium or copper creates an opportunity to obtain new materials with previously unknown electrical and functional characteristics. There are different approaches and works on using different methods to develop such materials are in progress and include the following:

- Chemical synthesis - Covetic type of atomic blend (blending by synthesis of liquid metal with graphene, under appropriate casting conditions),
- Electrochemical deposition of graphene on elements of copper or another material,
- Casting a mixture of liquid metal with graphene to a form facilitating further plastic processing to make wires with required characteristics (continuous-cast conductors as input material for further wire making can be the product of the process),
- Deposition of monoatomic carbon coats (graphene) using the CVD method on the surface of Al or Cu wires and their further plastic processing to form wires with required characteristics,
- Mechanical synthesis of Al powders coated with graphene or Cu powders coated with graphene to a form facilitating their further plastic processing to make wires with required characteristics.

2. RESEARCH METHODOLOGY

The base material used for the synthesis process of the analyzed composites was pure copper of the Cu-OF grade with chemical composition shown in **Table 2** and graphene manufactured by ITME in Warsaw.

Table 2 Chemical composition of copper for synthesis (wt. ppm). As a result of the casting process, 9 castings were produced under different casting conditions.

Material	Ag	As	Bi	Pb	Se	Sb	Te	Sn	Zn	Fe	Ni	S	O ₂	Cu
CuOFE	8	<0.3	<0.3	2	<0.3	<1	<0.4	<0.5	1.8	1.2	1.3	1.4	2.1	rem.

Figure 1 shows the images of the input materials. **Figure 2**, in turn, presents microscope scanning images of a multi-layer flake graphene that were used for the study.

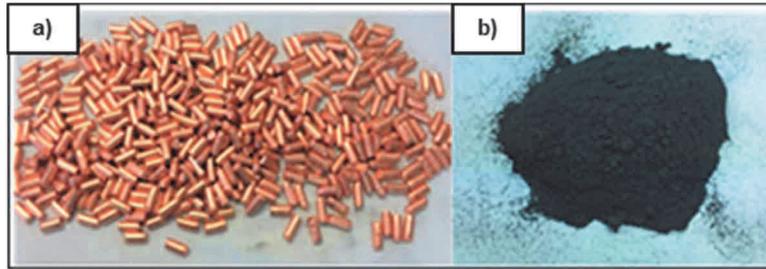


Figure 1 Material used for testing; a) copper Cu-OF, b) graphene manufactured by ITME

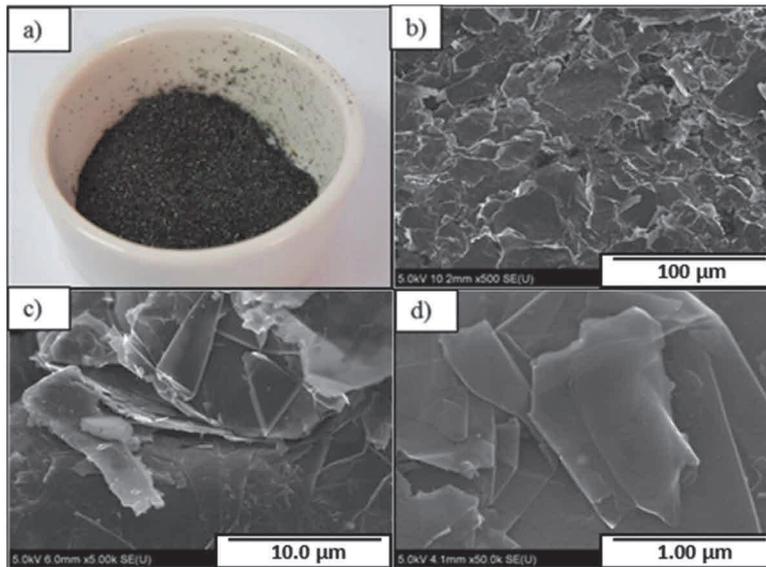


Figure 2 Summary of scanner images of graphene used for synthesis; a) the surface of the powder visible to the naked eye; b, c, d) various magnification

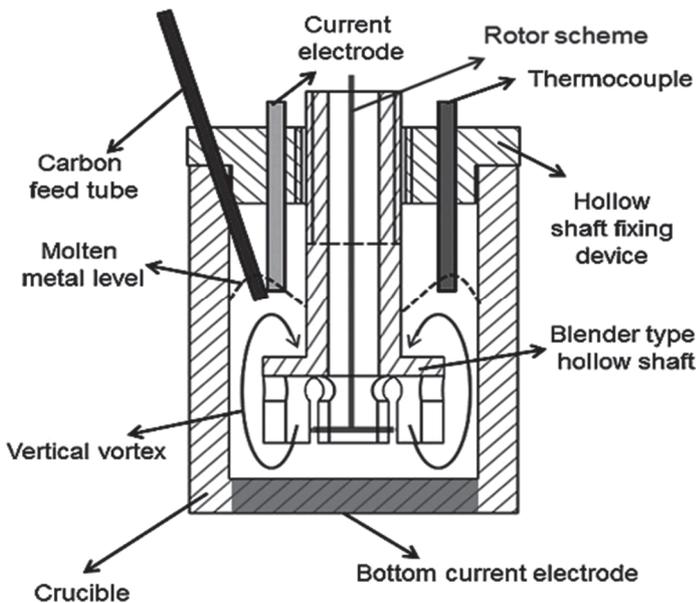


Figure 3 The concept and technical drawing casting furnace used for the synthesis of metallurgical copper graphene

The metallurgical synthesis of the copper-graphene composites was carried out on a specially designed continuous casting station shown in **Figure 1**. The layout of the test bench allowed the solving fundamental problems during the metallurgical synthesis of copper with graphene, i.e. the lack of wettability of graphene to copper and a three times lower density of graphene with respect to copper. These problems were solved by, e.g. the use of a special feeder system, which aimed to continuously dispense graphene into liquid copper directly into the crystallization zone. **Figure 3** shows the layout of the station.

The research consisted of a continuous casting process of a liquid copper-graphene mixture under various process parameters, i.e. different temperatures of liquid metal, different amounts of cooling water, different

pitch values and downtime during casting. In all cases, the same casting speed was used - 20 mm/s. Detailed parameters of the casting process are presented in **Table 3**.

Table 3 Parameters of the Cu-C composite casting process

Parameters of the casting process							
Casting No.	Coil strength in the furnace	Liquid metal temp.	Temp. of water cooling the crystallizer at the outlet	Cooling water flow	Casting temp. at the outlet	Pitch	Downtime
	(A)	(°C)	(°C)	(l/min)	(°C)	(mm)	(s)
1	300	1250	34	3.5	195	5	1
2	100	1330	40	1.9	205	5	1
3	300	1315	33	4.5	230	8	1
4	300	1230	32	7	215	4	0.5
5	300	1266	28	7	181	2	0.5
6	300	1270	30	6.8	245	5	0.5
7	300	1283	30	7	255	10	1
8	300	1296	27	10	185	5	1
9	100	1250	29	10	285	8	0.5

At a later stage of the study, castings with a diameter of 8 mm were tested for the effect of continuous casting parameters on physical properties, mechanical properties and casting structure. The mechanical properties test was performed on a Zwick v100 testing machine, the measuring base was 100 mm. Electrical conductivity test was made using the Thompson bridge. Surface roughness tests were performed using the contact method of a modern microprocessor-based profilograph on a Turbo Datwin-NT V1.4. Another study was to determine the density of a solid using a hydrostatic balance in a study based on Archimedes' law. In the next stage, attempts were also made to draw wire castings with a diameter of 3 mm. The process of drawing bars into wires was done using a laboratory draw bench. The wires were drawn at a speed of 0.2 - 0.3 m/s with the use of carbide-tipped workpieces. The operating angle of the working cone was $2\alpha = 18^\circ$, the lubricant being vegetable oil. Strain hardening was 85%.

3. STUDY RESULTS AND THEIR ANALYSIS

As a result of the casting process, 9 types of Cu-C composite castings were made, whose cross-sectional and longitudinal macrostructures are shown in **Figure 4**.

Based on the macrostructure images it was found that depending on the parameters of the casting process, the macrostructure of the cast bar varies. Increasing the rate of heat recovery during the casting process by increasing the amount of cooling water results in a finer structure (see microstructures 3 and 7 in **Figure 4**).

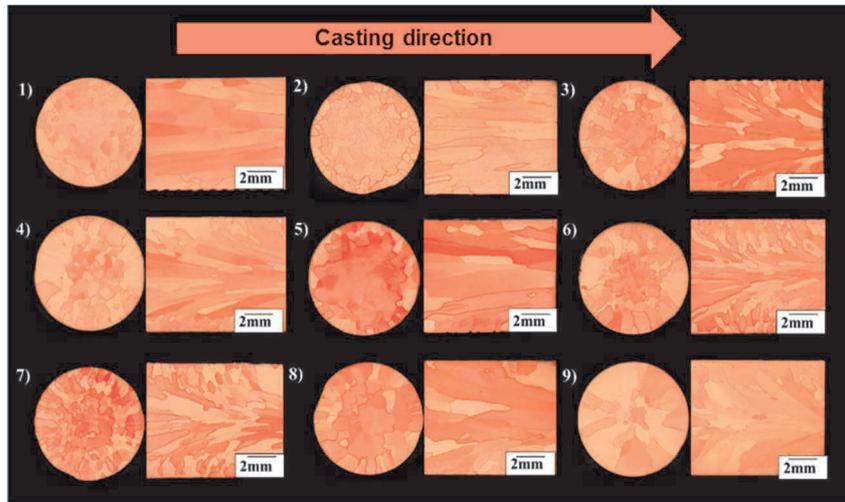


Figure 4 Macrostructure of obtained casts after the casting process, cross-section and longitudinal

In turn, reducing the intensity of cooling water results in a rough structure. Thus, the differentiation of process parameters in casting Cu-C composite leads to differentiation of the crystallization front which results in various cast structure. One of the primary issues is the efficiency of liquid copper and graphene synthesis. So the question is: Is graphene evenly distributed in copper and in what form is it present? To this end, microstructure studies were carried out using SEM techniques. The results are shown in **Figure 5**.

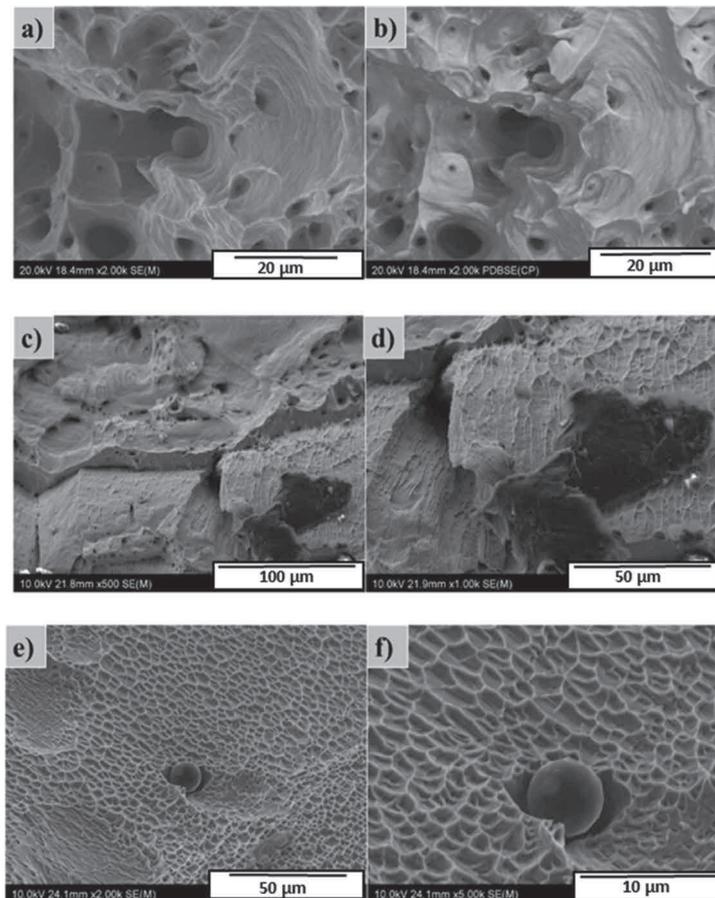


Figure 5 Scanning image of fractures after uniaxial stretching of selected Cu+C castings, showing forms where graphene was present

At a later stage of the study, the physical and mechanical properties of the bars were evaluated.

Figure 6 shows the results of electrical conductivity tests and the density of cast bars.

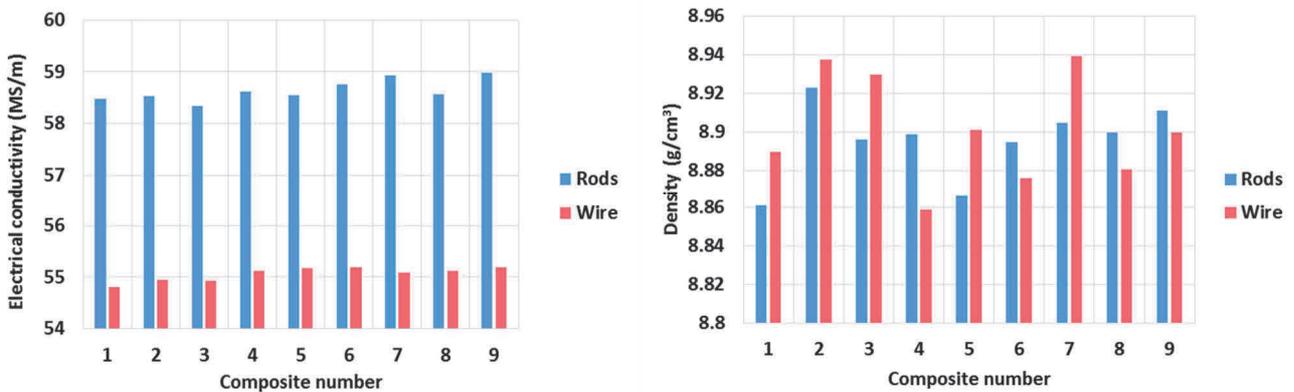


Figure 6 a) Comparison of the electrical conductivity for bars and wires obtained according to various casting parameters, b) Comparison of measuring the density of bars and wires received obtained by various casting parameters

Based on the tests, it was found that depending on the parameters of the casting process, the density of cast bars was 8.86 - 8.94 g/cm³. On the other hand, electrical conductivity tests did not show as much variation as in the case of density. In the next step, the bars were tested for mechanical properties, which were determined in a single-axis stretch test (see diagrams on the **Figure 7**).

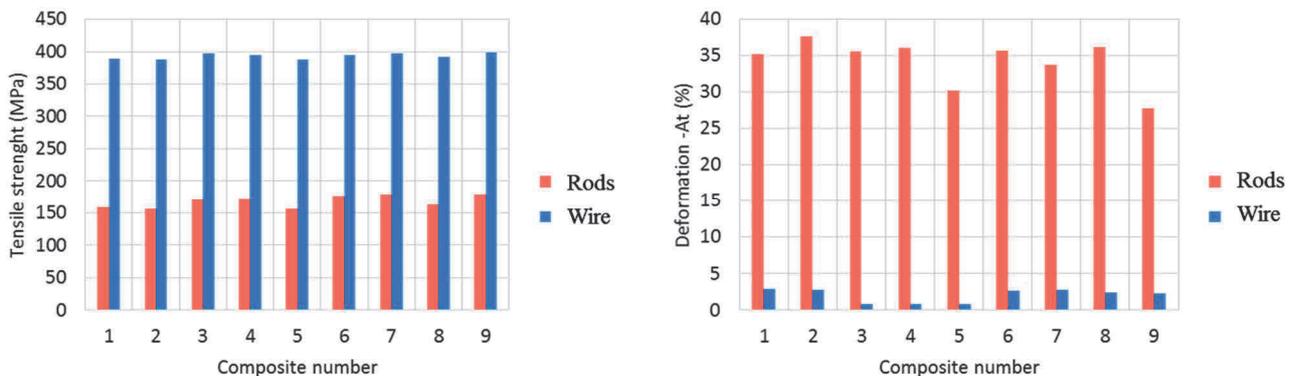


Figure 7 a) Summary of the tensile strength UTS for bars and wires of various obtained by various casting parameters, b) Summary of elongation parameters At for bars and wires obtained by various casting parameters

The tensile strength in all cases was similar and was about 150-170 MPa for bars and 400 MPa for wires. In turn, the total elongation of castings is varied by 27-37%. The drawing process of bars resulted in a significant decrease in plasticity, which was about 3% (see **Figure 7 b**).

The final stage of the research was the measurement of the surface quality. The results of the roughness are shown in the **Figure 8**. Difference in the roughness results of the bars results from the differentiation of the parameters of the casting process.

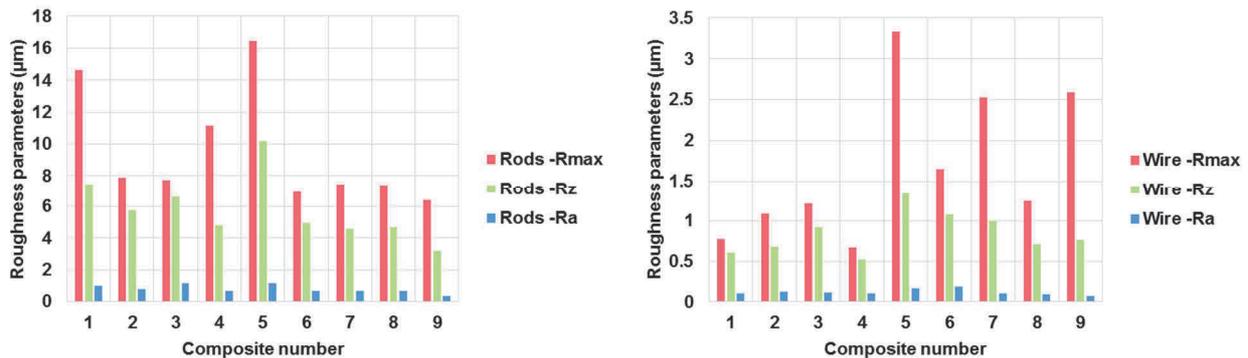


Figure 8 Summary of roughness parameters Rmax, Rz, Ra for bars and wires obtained by various casting parameters

5. CONCLUSION

Based on the study and the analysis of the obtained results, the following conclusions were made:

- The selection of parameters for the casting process within the range, which was carried out in the research, significantly influenced the macrostructure of the obtained castings. Due to the lack of wettability of graphene and copper and the lack of a significant difference in density of both materials, the coagulation process must take place as soon as possible. Hence, such conditions of the casting process (the amount of water and the type of feed) are preferred that guarantee rapid crystallization.
- The mechanical and electrical properties of the castings made are similar to the traditional Cu castings made by continuous casting. The only difference lies in the yield point, which is higher for Cu-C composites.

ACKNOWLEDGEMENTS

Authors are grateful for the financial support provided by The National Centre for Research and Development -Project No. PBS2/B5/30/2013 "New wires made of metal-graphene composites with increased electrical conductivity for OHL"

REFERENCES

- [1] TU, J. P., YANGA, Y. Z., WANGA, L. Y., MA, X. C., ZHANG, X. B. Tribological properties of carbon-nanotube reinforced copper composites. *Tribology Letters*, 2001, vol. 10, no. 4, pp. 225-228.
- [2] GUIDERDONI, Ch., ESTOURNE'S, C., PEIGNEY, A., WEIBEL, A., TURA, V, LAURENT, Ch. The preparation of double-walled carbon nanotube/Cu, composites by spark plasma sintering, and their hardness and friction properties. *Carbon*, 2011, vol. 49, no. 13, pp. 4535-4543.
- [3] KOLTSOVA, T. S., NASIBULINA, L. I., ANOSHKIN, I. V., MISHIN, V. V., KAUPPINEN, E., TOLOCHKO, O. V., NASIBULIN, A. G. New hybrid copper composite materials based on carbon nanostructures. *Journal of Materials Science and Engineering B*, 2012, vol. 2, no. 4, pp. 240-246.
- [4] NAYFEH, T. H., WIEDERHOLT, A. M. Nano-engineered ultra-conductive nanocomposite copper wire. Patent nr. US 2012/0152480.
- [5] RAO, C. N. R., MULLER, A., CHEETHAM, A. K. *The Chemistry of Nanomaterials*. Wiley-Vch, 2004.
- [6] DOLORES, A. M. *Trends in Nanotubes Research*. Nova Science Publishers, Inc. 2006.