Polish Society of Composite Materials
Polskie Towarzystwo Materiałów Kompozytowych

23: 2 (2023) 67-75

COMPOSITES THEORY AND PRACTICE
ISSN: 2084-6096
ISSN (online): 2299-128X

Dariusz Kołacz*, Joanna Kulasa, Karol Krukowski, Anna Hury, Grzegorz Muzia Małgorzata Osadnik, Jarosław Kalabis

Lukasiewicz Research Network – Institute of Non-Ferrous Metals, ul. Sowinskiego 5, 44-100 Gliwice, Poland
** Corresponding author: E-mail: dariusz.kolacz@imn.lukasiewicz.gov.pl*

Received (Otrzymano) 11.05.2022

EFFECT OF SINTERING TEMPERATURE AND ISOSTATIC PRESSURE ON SELECTED PROPERTIES OF SELECTED ELECTRICAL CONTACT MATERIALS MANUFACTURED BY HOT ISOSTATIC PRESSING

This paper presents the results of studies on the consolidation of metallic and composite powders by the hot isostatic pressing (HIP) process intended for electrical contact materials. Ag, Cu metallic powders, as well as AgW48Re2, AgRe1 and CuW47Re3 composite powders were used in the investigations. Green compacts for consolidation and hot sintering under pressure were prepared by double-sided axial pressing in steel dies. The density, electrical conductivity, and hardness were measured on the obtained sinters after the HIP process, and the microstructure was examined on selected ones. The research indicates that applying this technology allows good quality electrical material to be obtained, intended for electrical contact material.

Keywords: hot isostatic pressing, pressing, sintering, metallic powder, composite, sinter, contact material, hardness, density

INTRODUCTION

Progress in the design and production of new electrical devices creates the need to develop manufacturing methods to obtain materials for electrical contacts. The development of additive manufacturing technologies and devices for thermal and pressure processes (e.g., hot isostatic pressing – HIP) enables the production of connectors with complex shapes, often used for special applications.

When analysing the literature, attempts to use the above-mentioned additive manufacturing technologies to obtain silver or copper-based composites were observed, e.g., reinforced with metal oxides. In paper [1], the authors suggest the use of SLM technology for the production of Ag-based material with the addition of CuO. The Ag and CuO powders were subjected to mechanical synthesis and then laser-printed. The authors mention the porosity of the printed samples, which results in lowering the parameters of the current connectors.

One of the methods to improve the properties of such materials is to utilise the HIP process in the final stage of their production. The advantage of the hot isostatic pressing process is the possibility to eliminate internal porosity while maintaining the shape of the processed materials. The consolidation process uses an inert gas at high pressure (e.g., argon) as the pressure medium. The workpiece is simultaneously subjected to high isostatic pressure and high temperature. As a rule, powders sintered under pressure are in special capsules (cans, containers) designed for this purpose. The process of closing the powder in such capsules is commonly called canning. It is also possible to consolidate powders isostatically in the HIP process without special capsules, however, this technique requires their prepressing into the form of green compacts [2-9].

The application of the hot isostatic pressing process should improve the service life of connectors (especially those with complex shapes) produced by conventional powder metallurgy or, for example, by 3D printing.

The literature analysis reveals few works related to the use of the HIP process in the manufacturing of composite contact materials. One of the suggestions is to employ the HIP technique to produce electrical nanostructured W-Cu-Ni contact materials (mechanical synthesis + HIP) [10]. A different technology for producing contact materials was found in a patent [11], where the authors also indicate the possibility of obtaining the materials in the process of canning powders and densifying materials by HIP.

This article presents investigations of the densification of compacts (Ag, Cu, AgRe1, AgW48Re2, CuW47Re3) in the process of hot isostatic pressing produced by mixing and double-sided axial pressing in steel dies. Ag-W, Ag-Re and Cu-W composites were produced mainly by powder metallurgy techniques,

usually by infiltration of the tungsten preform with molten metal or high-energy milling in a planetary ball mill [12-17]. The presented scientific study is a proposal for the use of the hot isostatic pressing method in the process of manufacturing contact materials based on silver and copper, initially produced by conventional powder metallurgy.

RESEARCH METHODOLOGY

The density of the metallic powders was determined on an AccuPyc 1340 by Micromeritics. A Gemini 2360 by the same company was employed to test the specific surface area. The particle size distribution was measured on a Fritsch NanoTec Analysette 22 by the wet laser method (in water) in the measuring range from 0.10 μm to 504.48 μm according to the Fraunhofer theory.

The density of the obtained green compacts and sinters was determined using the geometric method. The electrical conductivity was measured by the eddy current method on a Foerster Sigmatest 2.069 instrument. HB hardness was determined using a 2.5 diameter ball applying a load of 612.92 N or 316.26 N for 15 seconds. Microstructural studies were performed on an Olympus GX71F metallographic microscope and a Zeiss Evo MA10 scanning electron microscope. Analysis of the phase composition was conducted by means of an XRD7 X-ray diffractometer by Seifert-FPM. Characteristic X-ray radiation Cu Ka and Ni filter were used. Dentification of the compounds was based on Seifert and Match software and ICDD PDF-4 catalogue data from 2022.

MATERIALS USED

Metallic powders of silver, copper, tungsten, and rhenium were used in this study. The Ag (> 99.99 wt.%), Cu (> 99.99 wt.%) and W (> 99.92 wt.%) powders were purchased from the Stanchem company. The Re powder was obtained by reducing ammonium perrhenate with hydrogen. Table 1 presents the chemical purity of the used ammonium perrhenate. The chemical composition of the metallic rhenium powder was not analysed.

TABLE 1. **Chemical composition of ammonium perrhenate**

Re	Zn	Mg	Cu	Mo	Ni	Pb	K	Na	Ca	Fe
wt.%	[ppm]									
69.45	3	< 3	< 3	< 5	< 5	< 5	5	4	< 3	< 3

Table 2 shows the properties of the powders and their characteristic points of particle size distribution. The actual density of the tested powders is lower than the theoretical due to the presence of microporosity in the particles. The development of the active surface of the powders is small, with the exception of rhenium for which the specific surface area is 1.314 m^2/g. The silver and copper powders have a similar particle size with an average particle size of about d50 = 23 μm, the tungsten powder is characterized by a slightly smaller size of particles (d50 = 13.87 μm), while rhenium is characterized by the smallest particle size (d50 = 1.07 μm).

TABLE 2. **Properties of powders used for studies and characteristic points of particle size distribution of metallic powders**

Metallic powder	Density [g/cm³]	Specific surface area – BET multipoint [m²/g]	Characteristic points of particle size distribution (d10, d50, d90)		
			d10	d50	d90
			[μm]		
Ag	10.32 ± 0.01	0.1549 ± 0.0028	7.81 ± 0.39	24.45 ± 0.73	58.39 ± 2.92
Cu	8.82 ± 0.01	0.3474 ± 0.0074	7.74 ± 0.39	22.94 ± 0.69	55.32 ± 2.77
W	19.11 ± 0.01	0.0477 ± 0.0039	5.22 ± 0.26	13.87 ± 0.42	32.66 ± 1.63
Re	19.73 ± 0.01	1.314 ± 0.0085	0.30 ± 0.02	1.07 ± 0.03	6.19 ± 0.31

Figure 1 presents the morphology of the investigated powders. The silver powder particles are characterized by a diversified shape, from spherical to globular to polyhedral particles. The particles of the copper powder are dendritic in nature. The tungsten powders come in the form of irregular polyhedron. The rhenium particles have an irregular shape, and their particles form large conglomerates.

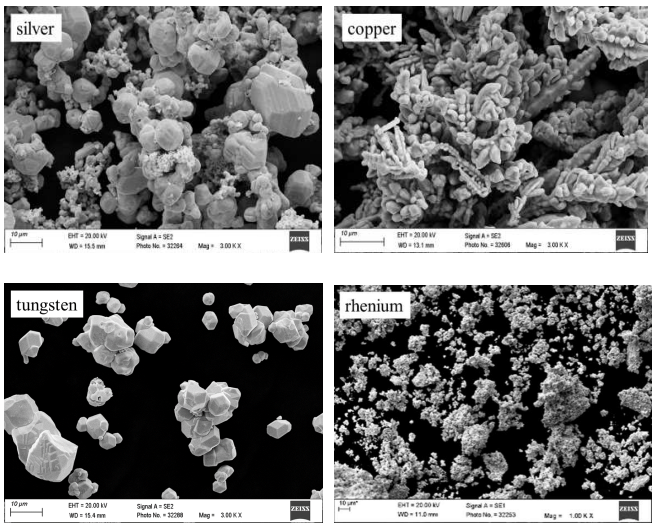


Fig. 1. Morphology of used powders

MANUFACTURING OF MATERIALS

The finished sinters were obtained by mixing the metallic powders, pressing and hot compaction in the HIP process. A schematic diagram of their manufacturing process is provided in Figure 2.

The chemical composition of the powders and powder mixtures for the studies is shown in Table 3. The mixtures were prepared by mixing the metallic powders in a bottle mixer for 1 hour.

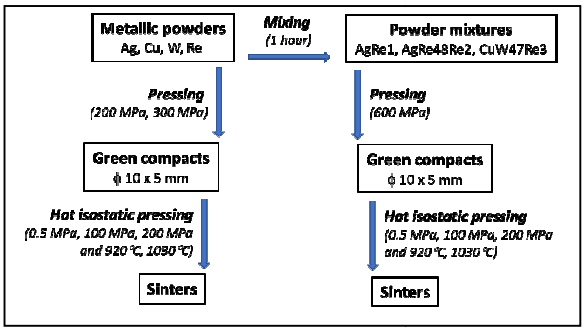


Fig. 2. Scheme of sinter production

TABLE 3. **Chemical composition of powder materials used for consolidation investigations**

	Ag	Cu	W	Re
	[% mass]			
Ag	100	-	-	-
AgRe1	99	-	-	1
AgW48Re2	50	-	48	2
Cu	-	100	-	-
CuW47Re3	-	50	47	3

All the powders and powder mixtures were pre-consolidated on a hydraulic press using double-sided axial pressing in steel dies to form green compacts with a diameter of 10 mm and a height of about 5 mm. The pre-pressing process to obtain the green compacts was conducted at 200, 300 and 600 MPa.

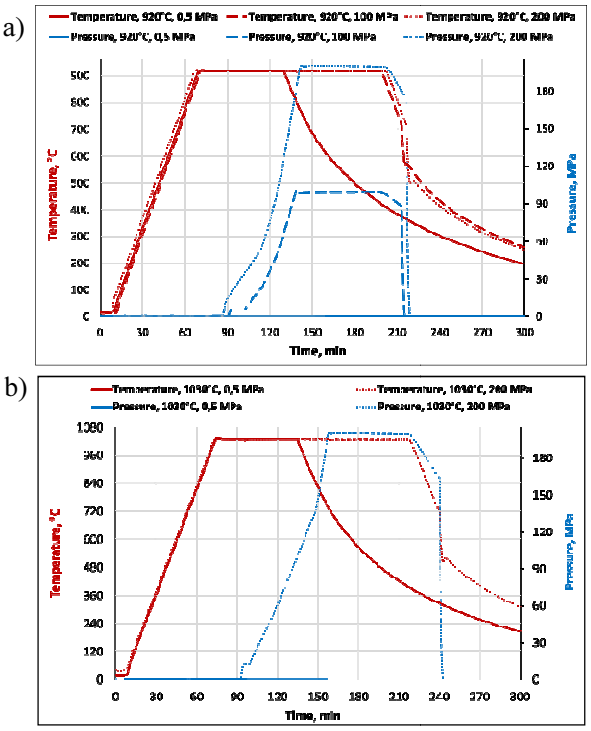


Fig. 3. Course of process curves of HIP process for green compacts consolidated: a) at temperature of 920°C, b) at temperature of 1030°C

The produced green compacts were hot isostatically consolidated using an AIP8-30H press from AIP American Isostatic Presses, Inc. The process, depending on the material, was conducted at different isostatic pressures (0.5, 100, 200 MPa) and temperatures (920 and 1030°C). Due to the purity of the process, a molybdenum furnace was used to sinter the green compacts. The HIP sintering process was carried out in two stages: in the first stage the green compacts were heated to the sintering temperature and sintered within 1 hour, while in the second stage a pressure gas was forced into the working chamber and the compacts were sintered again under isostatic pressure for 1 hour. The exception was the case in which a pressure of 0.5 MPa was applied. The samples were heated to the sintering temperature and then sintered for 1 hour and cooled in the furnace. The process curves are shown in Figure 3.

RESEARCH RESULTS

The density, electrical conductivity and HB hardness were measured on the obtained green compacts (pre-consolidation before the HIP process) and sinters after the HIP process. Microstructural investigations were carried out on selected samples (sinters after the HIP process). The results of the density and electrical conductivity measurements before are shown in Table 4 and after the HIP sintering process in Figures 4-7. The best results of plastic consolidation for the individual materials are presented in Table 5.

The results of selected microstructural observations are shown in Figures 8-11. One sample of each material was selected for analysis, considering its best plastic consolidation. The presence of pores was observed in the obtained sinters. Owing to the possibility of the formation of phases and chemical compounds during the HIP process (Figs. 10b, 11b), the phase composition of the materials analysed in this work was examined – Figures 12-14.

TABLE 4. **Properties of green compacts before HIP process**

Material	Pressing pressure[1] [MPa]	Density [g/cm^3]	Electrical conductivity [MS/m]	HB Hardness
Ag	200	8.67 ± 0.04	25.41 ± 0.33	31.8 ± 0.3
Ag	300	9.33 ± 0.06	32.83 ± 1.55	46.5 ± 1.1
AgRe1	600	9.86 ± 0.02	42.06 ± 1.24	70.1 ± 0.4
AgW48Re2		12.33 ± 0.04	17.53 ± 0.94	75.0 ± 2.0
Cu	200	6.37 ± 0.02	1.87 ± 0.24	28.4 ± 1.0
	300	7.05 ± 0.06	2.53 ± 0.28	45.1 ± 0.6
CuW47Re3	600	10.48 ± 0.04	1.67 ± 0.22	82.3 ± 1.8

[1] hydraulic press using double-sided axial pressing in steel dies

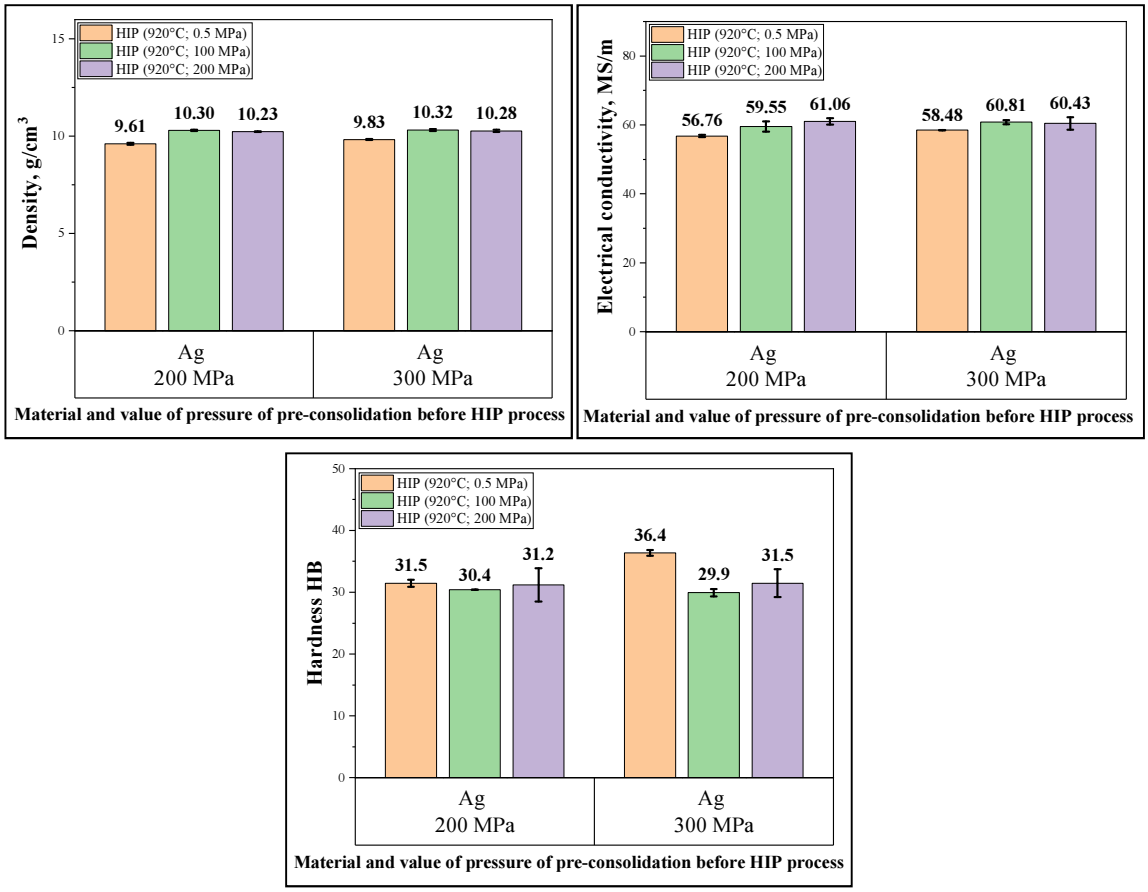


Fig. 4. Properties of silver sinters (after HIP process), 60 min

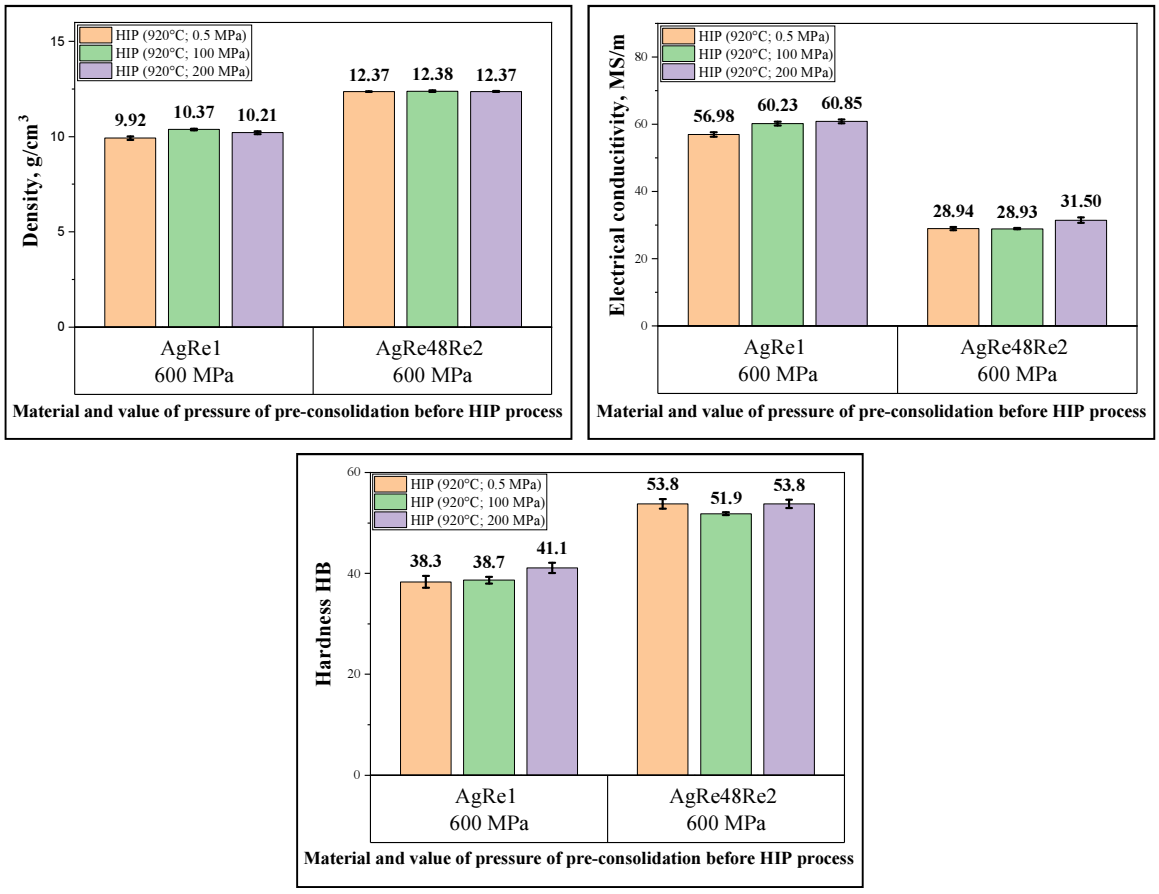


Fig. 5. Properties of silver-based sinters (after HIP process), 60 min

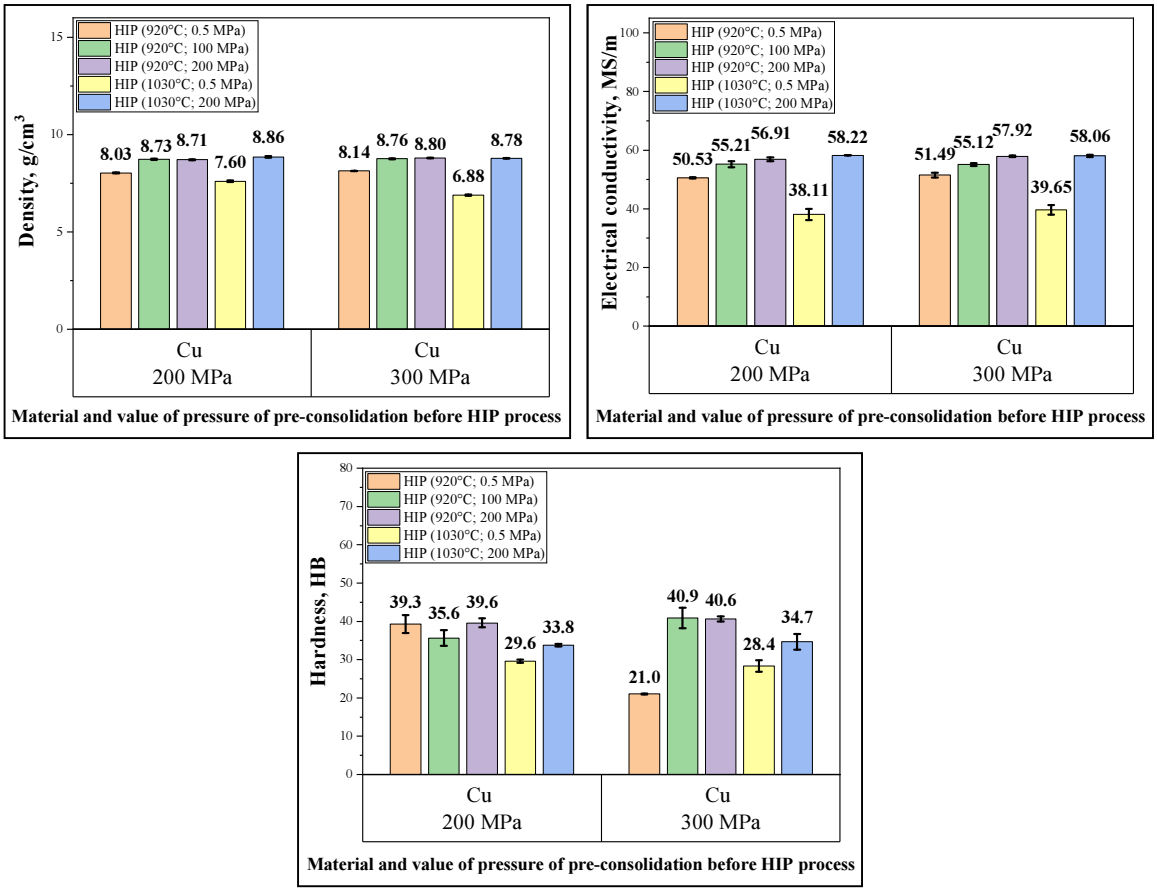


Fig. 6. Properties of copper sinters (after HIP process), 60 min

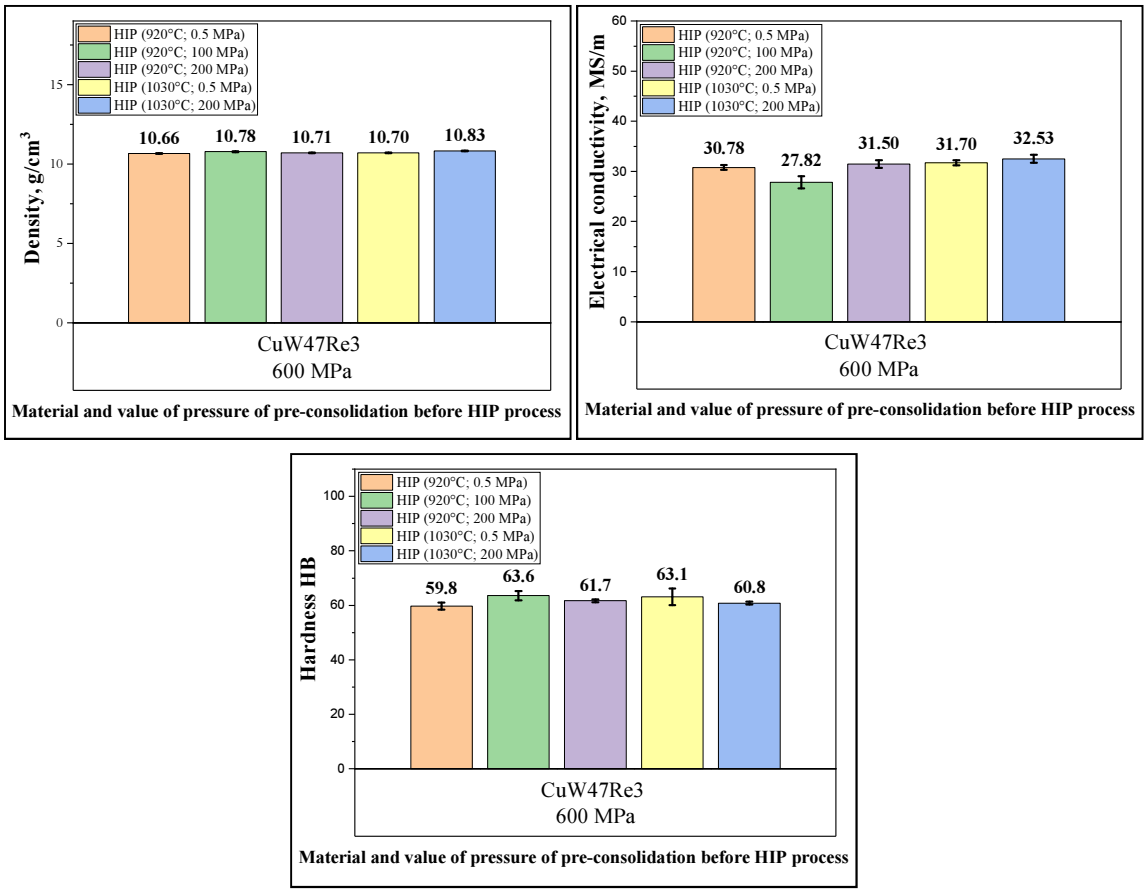


Fig. 7. Properties of copper-based sinters (after HIP process), 60 min

TABLE 5. **Properties of sinters for materials with best densification**

Material	Pressing pressure[1] [MPa]	Parameters of HIP process			Obtained parameters		
		Temperature [°C]	Isostatic pressure [MPa]	Time [min]	Density [g/cm^3]	Electrical conductivity [MS/m]	Hardness [HB]
Ag	300	920	100	60	10.32	60.81	29.9
AgRe1	600				10.37	60.23	38.7
AgW48Re2	600				12.38	28.93	51.9
Cu	200	1030	200		8.86	58.22	33.8
CuW47Re3	600				10.83	32.53	60.8

[1] hydraulic press using double-sided axial pressing

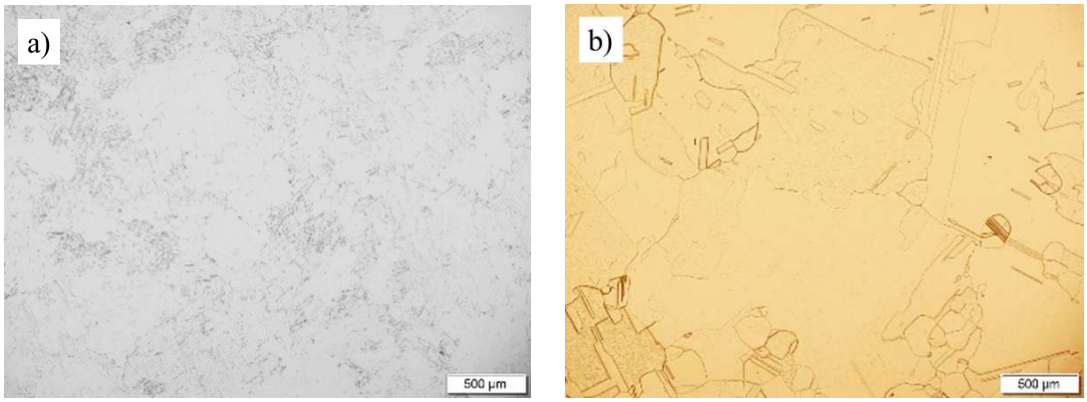


Fig. 8. Microstructure of sinters after HIP process, light microscope: a) Ag powder pre-pressed at pressure of 300 MPa, HIP 920°C, 100 MPa, 60 min, b) Cu powder pre-pressed at pressure of 200 MPa, HIP 1030°C, 200 MPa, 60 min

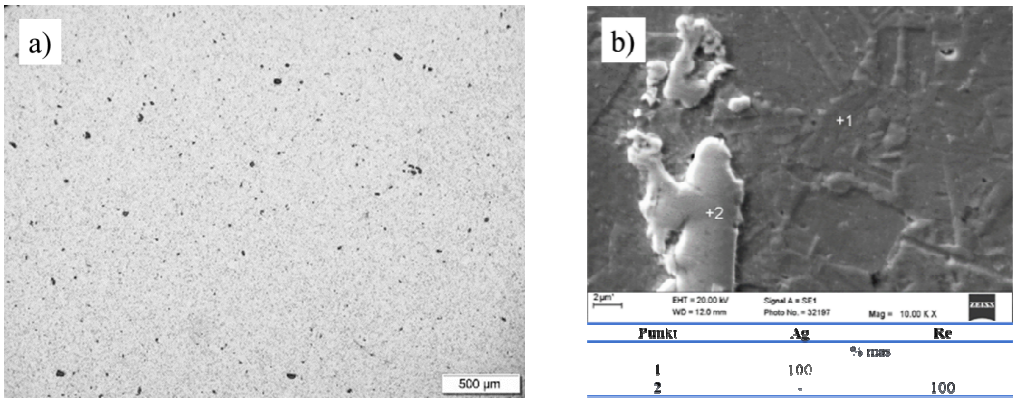


Fig. 9. Microstructure of AgRe1 composite, HIP: 920°C, 100 MPa, 60 min; a) light microscope, b) SEM

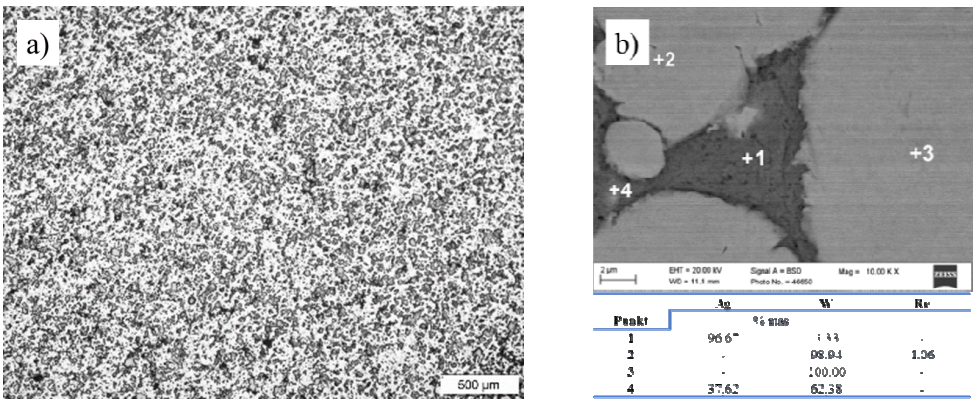
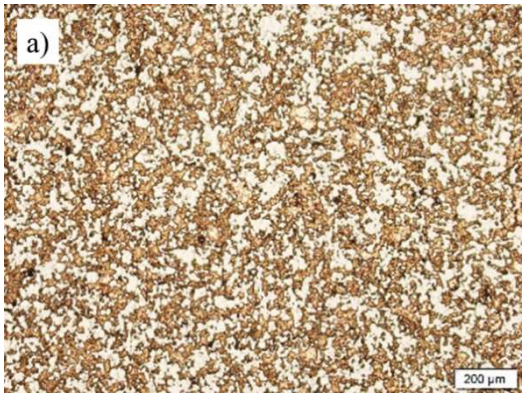
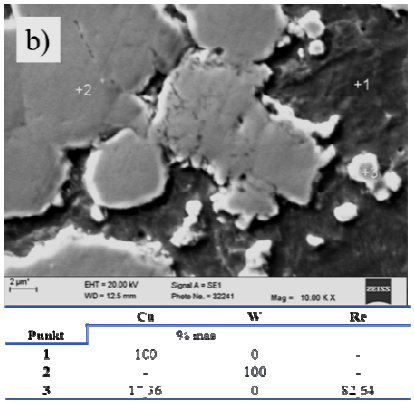


Fig. 10. Microstructure of AgW48Re2 composite, HIP: 920°C, 100 MPa, 60 min, a) light microscope, b) SEM





Punkt	Cu	W	Re
	% mas		
1	100	0	-
2	-	100	-
3	17.36	0	82.64

Fig. 11. Microstructure of CuW47Re3 composite, HIP: 1030°C, 200 MPa, 60 min; a) light microscope, b) SEM

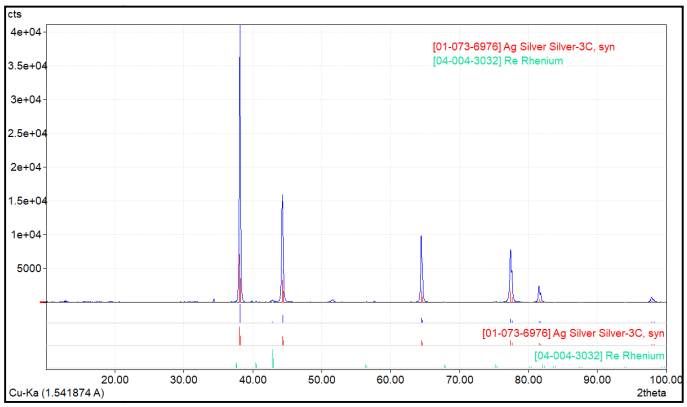


Fig. 12. Phase analysis of AgRe1 composite after HIP process, 920°C, 100 MPa, 60 min

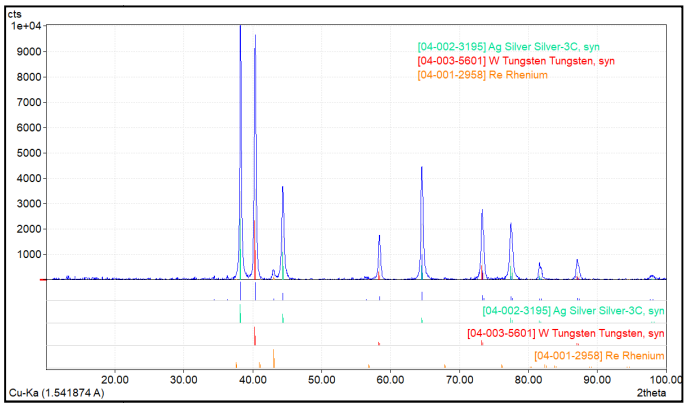


Fig. 13. Phase analysis of AgW48Re2 composite after HIP process, 920°C, 100 MPa, 60 min

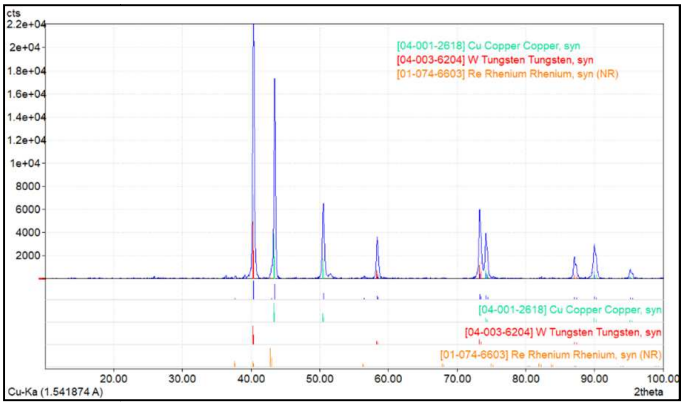


Fig. 14. Phase analysis of CuW47Re3 composite after HIP process, 1030°C, 200 MPa, 60 min

DISCUSSION OF RESULTS

For the hot isostatically pressed contact materials analysed in this paper, no similar scientific studies have been found, either in terms of the properties of the obtained materials or the parameters of the HIP process itself. The technology for the manufacture of the Cu-W-Ni material presented in [10] refers to a different chemical composition and to the manufacturing method itself in which, high-energy milling was additionally used prior to the HIP process.

Analysing the obtained investigation results for sinters made of pure silver, the highest density was obtained for the sample initially pressed with a pressure of 300 MPa and then hot isostatically densified at the temperature of 920°C and the pressure of 100 MPa. In the case of the AgRe1 composite, the highest density was also obtained using a lower isostatic pressure (100 MPa). However, for the AgW48Re2 composite, no significant effect of the applied isostatic pressure on the composite densification process was found.

The highest density of a sinter made of pure copper was obtained for the powder pre-pressed under the pressure of 200 MPa and then compacted in the HIP process using the temperature of 1030°C and the isostatic pressure of 200 MPa. In turn, in the case of the CuW47Re3 composite, the highest degree of sinter density was obtained at the temperature of 1030°C and the pressure of 200 MPa.

When analysing the microstructure of the sinters, discontinuities in their structure in the form of pores were noticed. The applied parameters of the hot isostatic pressing process (temperature, pressure, process time) did not enable their complete elimination. Changing these parameters, mainly by extending the process time, would probably improve the quality of the produced materials. The authors of [9, 18, 19] in their works used the time of proper HIP treatment equal to or longer than 2 hours.

The XRD study of the AgRe1, AgW48Re2 and CuW47Re3 composites does not indicate the formation of new compounds in the material during the HIP process (Figs. 12-14).

In addition, the previous use of the mechanical synthesis process and pressure infiltration before HIP post-processing would enable a significant increase in the properties of the investigated composites and extend their life in electrical connectors.

The obtained results indicate the possibility of using the HIP method in the production of contact materials, especially in cases where the shape of the final object cannot be changed, and it is necessary to increase its integrity, e.g., when using additive manufacturing technologies in the production of electrical materials [1].

CONCLUSIONS

The following conclusions were drawn during the research:

1. The presented technology of consolidating powders and powder mixtures in the process of hot isostatic pressing allows high-quality copper sinters to be obtained.
2. The initial pressing pressure of the powders and powder mixtures has a significant influence on the final properties of their sinters after the HIP process.
3. The best consolidation of sinters in the HIP process for the studied powder materials (Ag, AgW48Re2, AgRe1) was obtained using the temperature of 920°C and an isostatic gas pressure of 100 MPa as well as an initial pressing pressure for silver powder of 300 MPa, and for the AgW48Re2, AgRe1 powder mixtures initial pressing pressure of 600 MPa.

REFERENCES

[1] Chen S., Wang J., Yuan Z., Wang Z., Du D., Microstructure and arc erosion behaviours of Ag-CuO contact material prepared by selective laser melting, J. Alloys Comp. 2021, 860, 15894, 1-7, DOI: 10.1016/j.jallcom.2020.158494.

[2] Bocanerga-Bernal M.H., Hot isostatic pressing (HIP) technology and its applications to metals and ceramics, J. Mater. Sci. 2004, 39, 6399-6420, DOI: 10.1023/B:JMSC.0000044878.11441.90.

[3] Atkinson H.V., Davies S., Fundamental aspects of hot isostatic pressing: An overview, Metall. Mater. Trans. A 2000, 31, 2981-3000, DOI: 10.1007/s11661-000-0078-2.

[4] https://www.epma.com/epma-free-publications/product/introduction-to-hot-isostatic-pressing-brochure (access 25.04.2022).

[5] Wu J., Guo R., Xu L., Lu Z., Cui Y., Yang R., Effect of hot isostatic pressing loading route on microstructure and mechanical properties of powder metallurgy Ti_2AlNb alloys, J. Mater. Sci. Technol. 2017, 33, 172-178, DOI: 10.1016/j.jmst.2016.10.004.

[6] Hu B., Cai G., Fu J., Xin Y., Zhang D., Yuan Y., Guo S., Densification behaviour of tungsten alloy powders during hot isostatic pressing, Mater. Today Commun. 2022, 31, 103576, DOI: 10.1016/j.mtcomm.2022.103576.

[7] Kim H.G., Kim K.T., Densification behaviour of tungsten-fibre-reinforced copper powder compacts under hot isostatic pressing, Int. J. Mech. Sci. 2000, 42, 1339-1356, DOI: 10.1016/S0020-7403(99)00060-0.

[8] Hubler D., Ghasemi A., Riedel R., Fleck C., Kamrani S., Effect of hot isostatic pressing on densification, microstructure and nanoindentation behaviour of Mg-SiC nanocomposites, J. Mater. Sci. 2020, 55, 10582-10592, DOI: 10.1007/s10853-020-04758-5.

[9] Essa K., Jamshidi P., Zou J., Attallah M.M., Hassanin H., Porosity control in 316L stainless steel using cold and hot isostatic pressing, Mater. Des. 2018, 138, 21-29, DOI: 10.1016/j.matdes.2017.10.025.

[10] Tsakiris V., Lungu M., Enescu E., Pavelescu D., Dumitrescu G.H., Radulian A., Mocioi N., Nanostructured W-Cu Electrical contact materials processed by hot isostatic pressing, Acta Phys. Pol. A 2014, 125, 2, 348-352, DOI: 10.12693/APhysPolA.125.348.

[11] https://patentimages.storage.googleapis.com/3e/09/c9/b2b9f1b1735486/US4810289.pdf (access 20.12.2022).

[12] Madej M., Silver – based infiltrated composites, Archives of Metallurgy and Materials 2012, 57, 2, 605-612, DOI: 10.2478/v10172-012-0064-x.

[13] Wójcik-Grzybek D., Frydman K., Borkowski P., The influence of the microstructure on the switching properties of Ag-C, Ag-WC-C and Ag-W-C Contact Materials, Arch. Metall. Mater. 2013, 58, 4, 1059-1065, DOI: 10.2478/amm-2013-0126.

[14] Księżarek S., Woch M., Kołacz D., Kamińska M., Borkowski P., Walczuk W., Progress in fabrication technology of silver-based contact materials with particular account the Ag-Re and Ag-$SnO_2Bi_2O_3$ composites, Arch. Metall. Mater. 2014, 59, 501-508, DOI: 10.2478/amm-2014-0083.

[15] Kołacz D., Księżarek S., Woch M., Karwan-Baczewska J., Staszewski M., Rudnicki K., Borkowski P., Sienicki A., Nanocrystalline Ag-Re composite as a potential material for electric contacts fabrication, Arch. Metall. Mater. 2016, 61, 1847-1852, DOI: 10.1515/amm-2016-0298.

[16] Kołacz D., Księżarek S., Borkowski P., Karwan-Baczewska J., Lis M., Kamińska M., Juszczyk B., Kulasa J., Kowalski A., Wierzbicki Ł., Marszowski K., Jabłoński M., The influence of mechanical alloying and plastic consolidation on the resistance to arc erosion of the Ag-Re composite contact material, Materials 2021, 14(12), 3297, DOI: 10.3390/ma14123297.

[17] Matejicek J., Preparation of W-Cu composites by infiltration of W skeletons – review, Proceedings, 30th Anniversary International Conference on Metallurgy and Materials, Brno, Czech Republic 2021, May 26-28, DOI: 10.37904/metal.2021.4248.

[18] Hafenstein S., Werner E., Direct aging of a hot isostatically pressed A356 aluminium cast alloy, Mater. Sci. Eng. A 2019, 768, 138417, DOI: 10.1016/j.msea.2019.138417.

[19] Li J., Yuan Ch., J Guo, Hou J., Zhou L., Effect of hot isostatic pressing on microstructure of cast gas-turbine vanes of K452 alloy, Prog. Nat. Sci.: Mater. Int. 2014, 24, 631-636, DOI: 10.1016/j.pnsc.2014.10.008.

