INFLUÊNCIA DA TEMPERATURA DE SINTERIZAÇÃO E DA PRESSÃO DE COMPACTAÇÃO EM PASTILHAS DE COBRE PRODUZIDAS VIA METALURGIA DO PÓ

INFLUENCE OF SINTERING TEMPERATURE AND COMPACTION PRESSURE ON COPPER PELLETS PRODUCED BY POWDER METALLURGY

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Resumo: A metalurgia do pó é a técnica de fabricação de componentes metálicos por meio de compressão e sinterização de pós metálicos. O cobre tem sido usado por meio da metalurgia do pó para avanços tecnológicos e científicos devido às suas excelentes propriedades elétricas e mecânicas. Portanto, este estudo consiste em conhecer o comportamento e propriedades do pó de cobre puro na etapa de compactação e sinterização em pastilhas de cobre puro. Para a formação de pastilhas, a pressão de compactação foi variada entre 187,5 MPa até 700 MPa. A sinterização foi realizada em forno tubular com e sem controle de atmosfera com variação de temperatura, 700 °C, 800 °C e 900 °C. A pressão de compactação com 700 MPa proporcionou a obtenção de densidade relativa superior a 95%. A temperatura de sinterização e o controle de atmosfera mostraram que a variação nos valores da temperatura de sinterização tem influência significativa em diferentes resultados, como: densidade relativa, microdureza e microestrutura.

Palavras-chave: Metalurgia do pó. Cobre. Compactação. Sinterização.

Abstract: *Powder metallurgy is a manufacturing technique for metal components through the compression and sintering of metal powders. Copper has been used in powder metallurgy for technological and scientific advancements due to its excellent electrical and mechanical properties. Therefore, this study aims to understand the behavior and properties of pure copper powder during the compaction and sintering stages of pure copper pellets. For pellet formation, the compaction pressure varied between 187.5 MPa and 700 MPa. Sintering was performed in a tubular furnace with and without atmosphere control, with temperature variations of 700 °C, 800 °C, and 900 °C. A compaction pressure of 700 MPa resulted in a relative density higher than 95%. Sintering temperature and atmosphere control proved to be crucial for achieving better densification results.*

Keywords: *Powder Metallurgy. Copper. Compaction. Sintering.*

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I. INTRODUCTION

Copper has been widely used in the industrial and engineering fields due to its high electrical and thermal conductivity properties (Nazzer et al., 2019). Among the various metal manufacturing routes, powder metallurgy has a significant influence because it produces components with a high surface finish and strict dimensional control, as noted by Saberi et al. (2022). This method enhances material properties due to the arrangement of their internal structures, allowing for a wide range of applications in the industry of parts produced via powder metallurgy (Deepanraj et al., 2023).

Furthermore, understanding the variational parameters of compaction pressure and sintering temperature in this technique is essential, as they influence the material's microstructure and mechanical properties (Elkatatny et al., 2023). On one hand, compaction directly affects the geometric characteristics of the part, as highlighted by Pham et al. (2011). On the other hand, the temperature promotes solid-state diffusion bonding of the particles, resulting in controlled microstructure and mechanical properties of the compact, as pointed out by Vani (2018).

Powder metallurgy stands out from conventional metal production processes because it does not involve high temperatures, as in the melting process, and compaction can be performed at room temperature. It also eliminates the need for subsequent metal forming processes (Gohar et al., 2020). Moreover, by ensuring homogeneity and uniformity of the material's microstructure, this metallurgical technique results in products with high reproducibility and a high cooling rate (Deepanraj et al., 2023).

In Vincent et al. (2012), the effect of porosity volume fractions and their influence on the thermal conductivity of pure copper produced via powder metallurgy was studied. Another conclusion was that powder morphology and surface chemistry also impacted thermal conductivity (De Souza et al., 2021). Thus, understanding the behavior of pure metal in this manufacturing route is crucial for future applications, such as composites and nanocomposites.

Beyond being a manufacturing route for pure metals, powder metallurgy has gained prominence in recent years for its extensive use in producing metal matrix composites (Silva Junior et al., 2021). In composite production, where materials exhibit enhanced properties and are composed of more than one type of material, copper is the most studied and utilized matrix material in the industry (Usca et al., 2021).

Copper finds wide application in electronic components due to its excellent electrical and thermal properties. However, its low mechanical strength and wear resistance limit its application scope. Alloying copper with other elements is an alternative to enhance its strength, but this often results in a reduction in electrical conductivity (Akbarpour et al., 2023). Consequently, copper has been extensively studied regarding the improvement of its mechanical and electrical properties, whether in parts made of pure copper or copper-based metal matrix composites produced through powder metallurgy (Dixit & Srivastava, 2018; Wu et al., 2019).

Mastering the compaction and sintering parameters in powder metallurgy is crucial to processing a high-quality material with a high relative density, as well as optimized electrical and mechanical properties (Fermandes et al., 2023). Therefore, different parameters for compaction pressure and sintering temperature are employed depending on the composite being produced and whether the compaction is performed at room temperature or at elevated temperatures.

In the compaction phase, various pressures have been applied across studies. For cold uniaxial compaction, higher pressures are predominant, ranging from 400 MPa to 1 GPa, as observed in research by Ahmadein et al. (2021) and Swikker et al. (2020). On the other hand, for hot pressing, lower

pressures are used, such as 35 MPa, as seen in the studies by Wu et al. (2019).

Regarding the sintering process, which follows compaction, some studies utilize vacuum-controlled atmospheres, while others employ an inert argon atmosphere, with temperature ranges from 700°C to 1050°C (Usca et al., 2021; Swikker et al., 2020; Wu et al., 2019). In the analyses by Dixit and Srivastava (2018), pure copper pellets were sintered at 750°C without atmospheric control, leading to the appearance of surface pores. This allowed for an understanding of copper's behavior under conventional sintering conditions, opening possibilities for future studies.

As for the compaction and sintering of pure copper pellets, few studies have combined cold uniaxial compaction with conventional resistive furnace sintering. No studies were found comparing compaction pressure and sintering temperatures in a conventional atmosphere.

This work aims to analyze the influence of cold uniaxial compaction pressure at room temperature and sintering temperature on copper pellets. The goal is to broaden the discussion of powder metallurgy and its parameters, not only in the context of pure metallic materials but also for future applications in studies involving metallic composites and nanocomposites.

II. MATERIALS AND METHODS

The electrolytic copper powder used in this work was purchased from the company Metalpó Combustol and is of the PAC-2 type. According to the quality report provided by the company, this powder contains 99.5% pure copper with an average particle size of 20 μm.

The copper powder was analyzed using Optical Microscopy and Scanning Electron Microscopy (SEM). For metallographic analysis, 20 mm diameter pellets were cut using a Panambra cutting machine, model Mestotom. The material was mounted using a Struers mounting press, model Tempopress 2. After mounting, the samples were ground using a Struers grinder, model LaboPol-1. The optical microscopy for surface analysis was carried out using an Olympus microscope, model BX60M. SEM was used to analyze the geometry of the copper, graphite, and graphene oxide powders. The characterization was performed using a JEOL scanning electron microscope, model JSM-6510. The particle size and granulometry distribution of the copper powder were verified using a Tyler mesh sieve set: 150, 170, 250, 325, 400, and 500. The obtained results were then plotted on a particle size distribution curve.

The compaction procedure was carried out in the materials processing laboratory at Universidade Presbiteriana Mackenzie, using a Carver hydraulic press, model 3925, with a maximum load capacity of 22.7 tons. The operation involved loading approximately 12g of copper powder into an AISI D6 steel die with a 20 mm diameter cavity. To facilitate the process, the die was pre-coated with stearic acid to reduce friction between the tool and the compacted material. Cold uniaxial compaction was performed at varying pressures from 187.5 MPa (6 tons) to 700 MPa (22 tons) for 60 seconds to produce the composite pellets.

The sintering process was conducted in the laboratories of Universidad Presbiteriana Mackenzie. Sintering occurred a under controlled vacuum atmosphere using a horizontal tubular furnace, model FT-1700/H, with a maximum temperature of 1700°C. The sintering process began at room temperature and was conducted at target temperatures of 700 °C, 800 °C, and 900°C, with a 2 hours heating period. The sintering phase itself lasted for 1 hour, resulting in a total process time of 3 hours. After the sintering period, the furnace was turned off, and the samples were cooled inside the furnace, both with and without atmospheric control.

The density of the pellets is being calculated using Equation 1, with both sintered and green (only compacted) samples. This allows for the creation of a compressibility curve, enabling a comparative

analysis of these samples through relative density.

$$\rho_{rv} = \frac{\rho_a}{\rho}\ x\ 100 \qquad (1)$$

Where:

- ρ_a is the relative density of the samples (g/cm³);
- ρ is the density of copper (8,96 g/cm³).

The sintered samples underwent microhardness testing to aid in evaluating the evolution of the mechanical resistance of the composites due to changes in parameters during the experimental procedures. The tests were conducted in the materials processing laboratory at Universidad Presbiteriana Mackenzie, using a microhardness tester from Zwick/Roell, model ZHR8150CLK, equipped with a 50x objective lens and a diamond indenter under a load of 0.5 kgf for 10 seconds. A total of 25 random points were tested within each sample.

III. RESULTS AND DISCUSSION

The electrolytic copper powder used in this work was purchased from Metalpó Combustol and is of the PAC-2 type. Upon receipt of the copper powder, a chemical composition analysis was conducted using ICP-OES (Inductively Coupled Plasma Optical Emission Spectroscopy). The results of this analysis are presented in Table 1. The results obtained from the chemical analysis shows a similar outcome to the report provided by the responsible company.

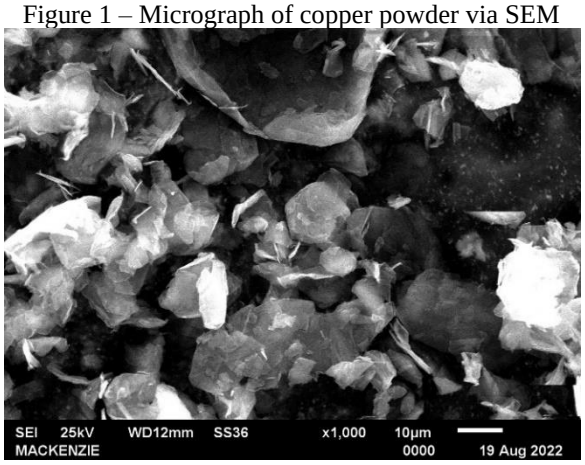
Table 1 – Chemical composition of copper powder.

Elements	(%)
Cu	99,5
Fe	0,3
Si	0,1
Zn	0,1

Source: Author,2024.

In the microscopy shown in Figure 1, the morphology of the copper particles can be observed. It is evident that the particles have an irregular shape, exhibiting a wide variation in size with a broad size distribution. This distribution consists of agglomerates of smaller particles, and the observed shapes are consistent with the electrolytic manufacturing process used and the report provided by the company from which the copper was purchased. Thus, the irregularity of the powder tends to contribute to the porosity of the material, while the presence of spherical particles enhances powder deformation during the compaction phase due to increased powder flowability (Jakub et al., 2016).

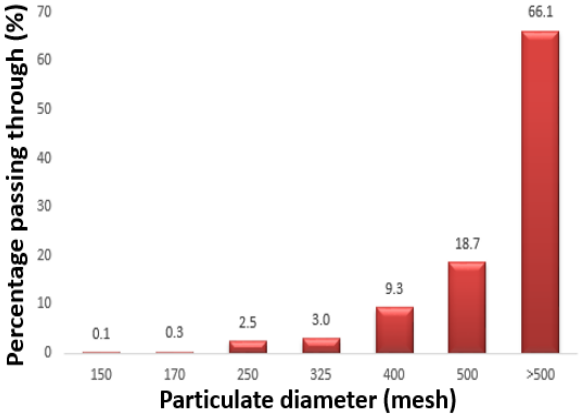
Figure 1 – Micrograph of copper powder via SEM



Source: Author,2024.

The granulometric distribution of the purchased copper powder was conducted to determine the particle size present. The results obtained are shown in Figure 2. It is noted that the predominant particle size (66.11%) exceeded the 500 mesh sieve, indicating that particles larger than 25 µm were present. Additionally, a wide granulometric range was observed, which contributes to a higher packing density (Huck-Jones, 2017).

Figure 2 – The granulometric distribution of copper powder



Source: Author,2024.

According to Faria (2017), compaction pressure is one of the most important parameters in the powder metallurgy process, as the density and porosity of a product manufactured through this method are regulated by compaction pressure. During the evaluation of compaction pressure with pure copper, compacting was performed at pressures of 187.5 MPa, 218.8 MPa, 250 MPa, 281.4 MPa, 312.7 MPa, 375 MPa, 437.5 MPa, 500 MPa, 600 MPa, and 700 MPa. The results obtained are shown in Table 2. Observing the pressures from lowest to highest, it is evident that the initial densification rate increases with increasing compaction pressure. According to Saboori et al. (2017), the significant increase in green density with the increase in compaction pressure can be attributed to the enhanced rearrangement of particles as pressure rises.

The compressibility curve was generated and is shown in Figure 3. It is possible to observe the densities obtained for the green samples as well as the densities after the sintering of the samples.

Observing the results, there is a continuous increase in compaction load; however, densification ceases to increase significantly, and an asymptotic behavior is observed in both the density of the green samples and the density of the samples after sintering.

In the work of Saboori et al. (2017), the asymptotic behavior of the density of the green compact is explained by the fact that at higher pressures, the increase in load no longer induces rearrangement of the particles. Consequently, beyond a certain point, there is a reduction in the volume of the samples.

Another observation is that with the increase in compaction load, the decrease in densification of the sintered samples leads to a significant increase in the density of the samples. However, at higher pressures, only a slight increase in density is observed after sintering. According to Nadkarni (1998), this behavior is expected, as samples with higher green density tend to exhibit less reduction when sintered. This is attributed to the lower degree of isolation of the channels and coalescence of the pores. Despite the smaller contraction, samples compacted at higher pressures continue to demonstrate greater density when sintered due to the higher density of the compact.

Table 2 – Chemical composition of copper powder.

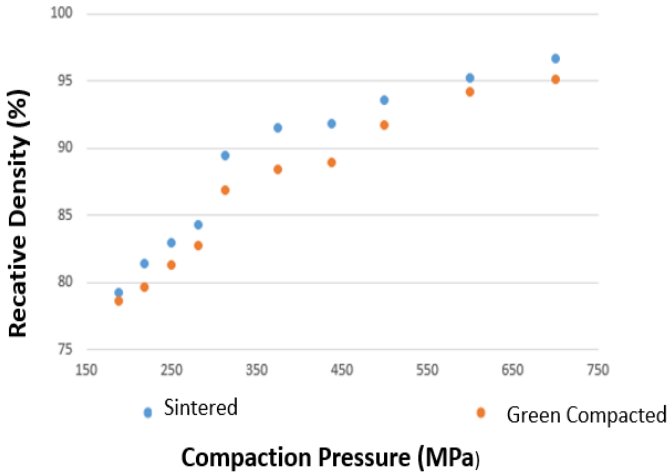
Compaction Pressure (MPa)	Relative Density (%)
187,5	79,2 $^{\pm 0,6}$
218,8	81,4 $^{\pm 0,7}$
250	82,9 $^{\pm 1,0}$
281,4	84,3 $^{\pm 1,0}$
312,7	89,4 $^{\pm 1,4}$
375	91,5 $^{\pm 1,4}$
437,5	91,8 $^{\pm 1,8}$
500	93,6 $^{\pm 2,1}$
600	95,2 $^{\pm 1,5}$
700	96,7 $^{\pm 1,2}$

Source: Author,2024.

The Vickers microhardness values of the sintered samples were measured, and the average values obtained are shown in Table 3. In the tests conducted, the pressure of 700 MPa yielded the best results in the densification of the samples sintered at 900 °C, achieving a Vickers microhardness value of 96.7% with an average Vickers microhardness of 57.21 HV.

Therefore, it was decided to continue working with the pressure of 700 MPa in the evaluation stage of the sintering temperature.

Figure 3 – Relative Densities of Green and Sintered Samples of Pure Copper.



Source: Author, 2024.

Table 3 – Average results obtained for the evaluation of mass values after sintering

Compaction Pressure (MPa)	Vickers Microhardness (HV)
187,5	33,9 ± 2,8
218,8	35,9 ± 3,9
250	35,7 ± 2,8
281,4	39,9 ± 2,1
312,7	42,9 ± 2,5
375	44,6 ± 2,4
437,5	46,2 ± 3,0
500	50,9 ± 2,0
600	52,6 ± 2,2
700	57,2 ± 2,4

Source: Author,2024.

With the definition of the compaction pressure, a study was conducted on the variation of sintering temperature. Based on the literature, three sintering temperatures were selected: 700 °C, 800 °C, and 900 °C. The values obtained from the tests are shown in Table 4.

Table 4 – Average results obtained for the evaluation of relative density for samples at different sintering temperatures.

Compaction Pressure (MPa)	Vickers Microhardness (HV)
700	92,7 ± 2,8
800	93,9 ± 2,4
900	96,7 ± 1,1

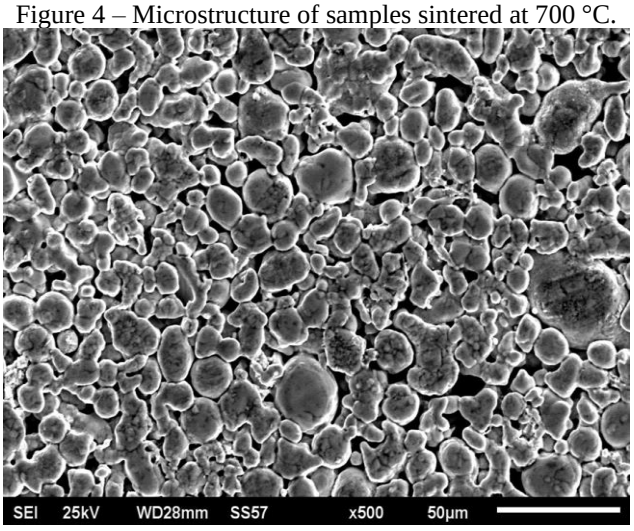
Source: Author,2024.

Figure 4 shows the microstructure of the sample sintered at 700 °C, indicating that there was not a complete sintering of the samples, as voids are observed.

Figure 5 presents the microstructure of the sample sintered at 800 °C, where improved sintering behavior was observed compared to the sample sintered at 700 °C. Analyzing the figure, the agglomeration of the powder particles is evident.

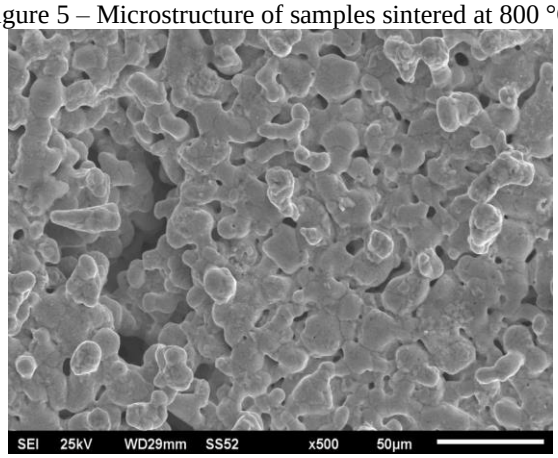
Figure 6 presents the microstructure of the sample sintered at 900 °C, where a greater degree of particle sintering is evident, with the particles being fully melted compared to Figure 5. There is a noticeable decrease in the number of voids and an increased consistency in the agglomeration of the powder particles.

Figure 4 – Microstructure of samples sintered at 700 °C.



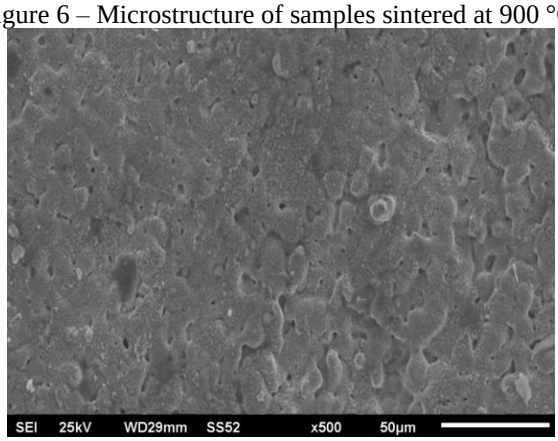
Source: Author, 2024.

Figure 5 – Microstructure of samples sintered at 800 °C.



Source: Author, 2024.

Figure 6 – Microstructure of samples sintered at 900 °C.



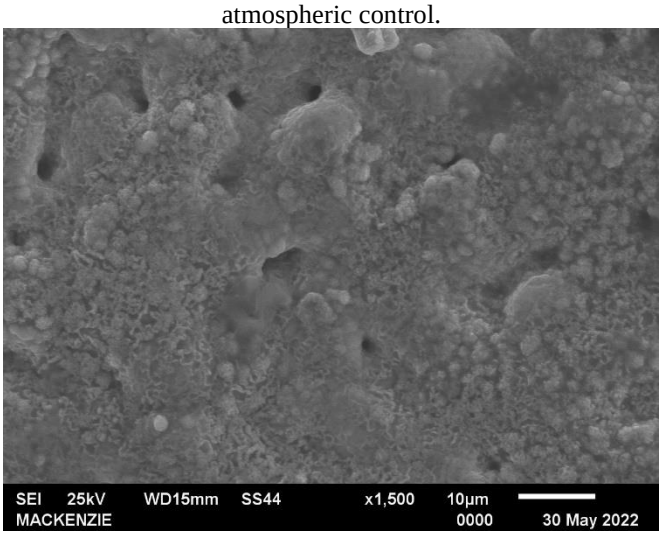
Source: Author, 2024.

Based on the literature, lower temperatures result in slower diffusion of atoms during sintering, allowing for the internal pores to close before the outer parts of the samples. This decreases the retention of expandable gases inside the samples, ultimately leading to a reduction in the dimensions of the samples.

Another point evaluated was the control of atmosphere, considering the conditions available in the equipment belonging to the laboratories of the Presbyterian University of Mackenzie. Two scenarios: with nitrogen atmosphere control and without atmospheric control.

Figure 7 shows the microstructure without atmospheric control, where oxidation of the pellet is clearly visible, confirming the necessity of atmosphere control during the process to prevent the formation of copper oxide.

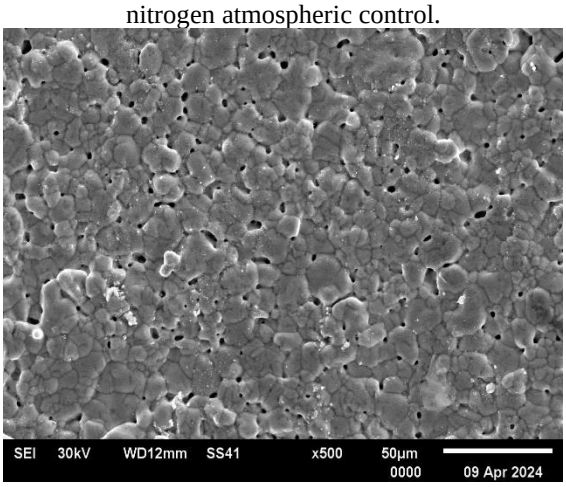
Figure 7 – Microstructure of samples sintered at 900 °C without atmospheric control.



Source: Author, 2024.

In Figure 8, it can be observed that there was no oxidation of the pellet; however, the formation of pores occurred due to gases trapped during the process, leading to a reduction in the mechanical properties of the pellet.

Figure 8 – Microstructure of samples sintered at 900 °C with nitrogen atmospheric control.



Source: Author, 2024.

Upon completion of the sintering processes, Vickers microhardness tests were conducted, as shown in Table 5. The evolution of the microhardness of the samples with atmosphere control is evident, along with the improvement in the microstructure.

Table 4 – Average results obtained for the evaluation of relative density for samples at different sintering temperatures.

Condition	Vickers Microhardness (HV)
Without control	35,8 $^{\pm 1,8}$
Nitrogen	43,6 $^{\pm 1,4}$

Source: Author,2024.

IV. CONCLUSIONS

The compaction pressure of 700 MPa resulted in a relative density exceeding 95%. The optimization of the sintering temperature proved to be essential for achieving better results in densification; therefore, a temperature of 900 °C was utilized. Atmosphere control also proved to be crucial, and vacuum was employed during the process. The Vickers microhardness measured was 43.6 HV, with the best results obtained from samples sintered under nitrogen atmosphere control.

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