Effects of grading tungsten powders on properties of CuW alloy

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Abstract
Considering the advantages and disadvantages of CuW alloys with different single particle size of tungsten powder, CuW alloys with three kinds of tungsten powders: ultramicron (50 μm), micron (6–8 μm), submicron (0.4 μm) were prepared by the infiltration method. Microstructure of the CuW alloy with different grading ration of tungsten powder were characterized via field-emission scanning electron microscope (SEM). The Vacuum electrical breakdown properties were studied by electrical breakdown test. The results shows that the grading tungsten powders can form a variety of W-W sintered necks, strengthen the tungsten skeleton. SEM results show that a suitable ratio of grading tungsten powders can make the infiltrated copper phase more dispersed and uniform. Compared with the tradition CuW alloy, the conductivity is greater than 53.5%IACS, with the lowest hardness of 203HB, the lowest CuW/CrCu bonding strength of 417 MPa and the highest of 495 MPa, the hardness and electrical conductivity increased by 20%–30%, the bonding strength of new CuW/CrCu monolithic materials increased by 20%, vacuum electrical breakdown properties performed good.

1. Introduction

CuW alloys are widely used in various circuit breakers and transformer switches due to their good electrical properties, high thermal stability and good resistance to arc erosion [1–3]. Nowadays, with the development of production, increase the capacity of the new type of high voltage circuit breaker, the arc extinguish chamber space is narrow, so as to make the arc contact per unit area of load increase, on the whole contact material performance put forward higher requirements. CuW alloy prepared by conventional single particle size of tungsten powder has been difficult to meet the requirements of current electrical contacts in practical applications. Considering the advantages and disadvantages of CuW alloys with different single particle size of tungsten powder. The CuW alloy prepared with submicron tungsten powder has high hardness and electrical conductivity, the average value was found to be about 201HB, the conductivity was 57%IACS, which was close to the alloy prepared by the grading method and the Cu matrix has good continuity and uniformity. However, due to the submicron powder is too fine, the specific surface energy is high, and the apparent density is small. Therefore, the process for preparing the alloy is extremely cumbersome, and the powder is difficult to be compact with conventional cold-pressing method, the impurities in the air are more likely to be brought into during the pressing process and seriously affect the performance of alloy, which will destroy the continuity of the tungsten skeleton and greatly reduce the conductivity and other properties of the alloy [4]. The microm tunnel powder is relatively easy to handle, and the process for preparing the CuW alloy is relatively simple. It is the most widely used and most common CuW alloy at present. But the vacuum electrical breakdown performance and overall strength are not the best [5]. The CuW alloy prepared with ultramicron tungsten powder has high strength and good thermal stability, the lowest strength up to 460MPa, but the continuity of matrix is too poor to produce copper-rich phase, and due to its high apparent density, the density of prepared CuW alloy is too high to be utilized in practical applications [6–8].

In our previous study [4], particle size have a great influence on bonding strength and bulk density. Ultramicron tungsten powder particle size is larger, the bulk density is small, so the specific surface area is lesser, grain boundary is less, when fracture crack through the large grain size, the strength of the alloy showed good,
micron and submicron powder grain size is small, the bulk density is larger, larger specific surface area, the grain boundary is more, when fracture cracks along the grain boundary extension, alloy strength is poorer. It can be seen that a reasonable grading ratio can obtain the best molding density, the most reasonable number of sintering necks, and the number of sintering necks will directly affect the strength of the material [7, 9, 10].

In this study, the experiment wanted to improve the performance of CuW alloys by adjusting the tungsten powder grading ration to increase the number of different W-W sintering necks. The CuW alloy composition design scheme and diagram of tungsten skeleton structure shown in figure 1. The skeleton structure and the distribution of Cu-W two phases are changed by different grading schemes. The results demonstrated a more convenient way to obtain CuW alloys with a more desired uniform microstructure, higher hardness, higher conductivity and better Vacuum electrical breakdown properties.

2. Experimental

According to the designed powder schemes, as shown in table 1, weighed different powders (submicron, micron, ultramicro tungsten powders and induced Cu powders) under various grading schemes, then put each group of mixed powders and the steel mill balls with the ration of 1:1 into the GM-D/B type mixing machine. Mixed the powders for 4 h at 100 r min⁻¹ speed. Then sprayed the mechanically mixed powders with wax, dried at room temperature and sieved. According to the preset compaction ratio of CuW70 alloy, weighed a certain amount of mixed powder and put it into the mold, distributed the powder uniformly in the mold cavity by knocking and vibrating mold sleeve to ensure in order to ensure that the compression ratio of each part of the blank is consistent. The compact was pressed at the 300 KN pressure for 30 s with diameter is Φ24mm. After the pressure is removed, took out the compact from the moulding-die. In this experiment, the density of compact is 10.05 g cm⁻³. The W skeletons were firstly sintered under hydrogen atmosphere at 1000 °C for 2 h, followed by putting pure copper block (purity N99.9%) on the sintered W skeleton in a graphite crucible, and infiltrating at 1350 °C for 2 h under hydrogen atmosphere. Then obtain CuW alloy having a predetermined composition. Subsequently, the prepared CuW sample and the CrCu alloy are integrally sintered under the protection of hydrogen to obtain the CuW/CrCu monolithic material. Finally, The treatment temperature of CuW/CrCu was 1000 °C–1050 °C, the treatment time was 1.5 h, the aging temperature was 440 °C–460 °C, and the aging time was 4 h.

The microstructure morphology of CuW alloy was examined in Oxford JSM-6700F field emission scanning electron microscope (SEM). The electrical breakdown was tested in an arc extinguishing chamber modified by a

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**Table 1.** Composition design of CuW alloy with grading tungsten powder.

| The content (%) of 0.4 μm W powder in the total W powder | W(0.4 μm) | W(6 ~ 8 μm) | W(50 μm) | Cu |
|----------------------------------------------------------|-----------|-------------|-----------|----|
| 13%                                                      | 10.5      | 49          | 10.5      | 30 |
| 20%                                                      | 14        | 42          | 14        |    |
| 30%                                                      | 21        | 28          | 21        |    |
| 40%                                                      | 28        | 14          | 28        |    |
| 45%                                                      | 31.5      | 7           | 31.5      |    |

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Figure 1. Diagram of tungsten skeleton structure and the design diagram of CuW alloy composition.
time and other discharge parameters. The formula of breakdown strength is as follows:

\[
E = \frac{U}{D}
\]

Then investigated other basic properties, including the density, hardness, electrical conductivity, and properties of CuW/CrCu monolithic materials, analyzed the regularity of various performances changed with the tungsten powder grading.

3. Results and discussions

3.1. Morphology of three kind tungsten powders

Figure 2 is the SEM micrographs of submicron (0.4 μm), micron (6 ~ 8 μm) and ultramicron (50 μm) tungsten powders magnified 2000 times. The submicron tungsten powder has small particle size, large degree of dispersion and specific surface area. On the contrary, the ultramicron tungsten powder has large particle size, and the degree of dispersion and specific surface area are small. For liquids, the surface Gibbs function, \( \Delta G_s = \sigma A_s \), where \( A_s \) represents the surface area, and \( \sigma \) is the proportional coefficient. It can be seen from the above formula, the greater the surface area of liquids, the higher the surface energy. For solids, due to the absence of adjacent atoms around the surface atoms, there exist many empty dangling bonds with unsaturated property, which lead to these surface atoms have high chemical activity.

For powder materials with large specific surface area, the influence of surface energy on their properties cannot be ignored. The specific surface area of the powder material is inversely proportional to the size of its single particle. For the three particle size tungsten powders used in the experiment, submicron (0.4 μm), micron (6 ~ 8 μm) and ultramicron (50 μm). The ratio of its specific surface energy and its specific surface area is approximately 8:4:1.

3.2. Microstructure of CuW alloy

As shown in figures 3(a) to (e) are microstructures of CuW alloys magnified 200 times under five different tungsten powder grading ration. From (a) to (e), it can be clearly seen that the content of ultramicron (50 μm) tungsten powder and content of submicron (0.4μm) tungsten powder increases, while the content of micron powder (6 ~ 8 μm) decrease, this is consistent with the composition designed. It can be seen that the skeleton structure of CuW alloy prepared by this tungsten powder grading method is continuous, and the tungsten phase and the copper phase are uniformly distributed, and copper enrichment is rarely present.

The darker place in figure is the copper phase and the brighter place is the tungsten phase. The three different tungsten powders used in this experiment are all orthorhombic structure. Although their particle size is different, their crystal structure is the same. So the structure of tungsten powder has little influence on the performance of CuW alloy prepared by grading method in this experiment and can be ignored.

Enlarge the microstructure of figure 3(c) in different multiples to get the figure 4. It can be seen from the figure 4 that the grading tungsten powders can form a variety of W-W sintered necks. The dispersion of ultramicron tungsten powder is relatively uniform, and there is almost no contact between the ultramicron tungsten powder, so it is difficult to form equal-diameter sintered necks between ultramicron tungsten powders. The content of micro powder is large, and there are more opportunities for them to contact with each other. It is easy to form equal-diameter sintered necks of micron powder, and its sintered neck is relatively large and obvious. Submicron powders are aggregated together due to their agglomeration, and the equal-diameter sintered necks are relatively obvious and small. In addition, it can be seen from figure 4 that non-equal diameter sintered necks
are formed between three different kinds of tungsten powders. The ultramicron tungsten powder is difficult to be combined with other powders due to its high thermal stability, so the non-equal diameter sintered necks are formed less. While the micron-sized tungsten powder and the submicron tungsten powder have higher surface energy and lower thermal stability, so more good non-equal diameter sintered necks are formed between them. This structure would strengthen the tungsten skeleton and improve alloy properties.
3.3. Hardness and electrical conductivity

Deng et al. [11] studied the CuW composite coating material, and the average hardness was about 200HV. Chen et al. [12] proposed that the hardness of CuW70 alloy prepared by traditional methods was about 187 ± 13 HV, while they raised the hardness of CuW70 alloy prepared by microwave sintering method to 204 ± 13 HV, which was lower than the minimum hardness of the alloy we prepared. Figure 5 is the hardness and electrical conductivity curve of CuW alloys with different tungsten powders grading scheme. It can be seen from figure 5 that the hardness and electrical conductivity of the CuW alloy prepared by this tungsten powder grading method are both higher. The CuW alloy prepared by this method breaks the regularity that the electrical conductivity decreases with the hardness increases of the conventional CuW alloy. It can be seen from the figure that the electrical conductivity of the CuW alloy increases with the increase of the submicron tungsten powder content. The hardness of CuW alloy is at the highest value when the content of submicron, micron and ultramicron tungsten powders are 40%, 20% and 40% respectively.

The increase of the hardness of CuW alloy is due to the increase of ultra micrometer tungsten powder, while the increase of electrical conductivity of CuW alloy is due to the increase of the submicron tungsten powder. Ultramicron tungsten powder has high strength and high hardness. The addition amount in a certain range will greatly increase the strength of the alloy skeleton, thus improved the strength and hardness of the whole CuW alloy. The submicron tungsten powder has a large specific surface area. During the sintering and infiltration process, the copper phase is uniformly dispersed around it, which greatly increases the specific surface area of the copper phase in the alloy and the dispersion and continuity of copper phase. Finally improve the electrical conductivity of CuW Alloy. The smaller the particle size of tungsten powder, the higher the degree of dispersion of copper in the alloy, and the less likely to be enriched.

The ratio of the single particle surface area of three tungsten powders used in the experiment was 15000:200:1, and the ratio of the total surface area of the particles in the same volume of the stack was 1:8:125 in an ideal condition. Therefore, the smaller the particle size of the tungsten powder, the greater the specific surface area of the high-conductivity copper around it. That is the reason why the submicron tungsten powder can improve the electrical conductivity while the ultramicron tungsten powder can reduce the electrical conductivity.

3.4. Vacuum electrical breakdown properties

Figure 6 is the SEM micrographs of the five CuW alloys after 50 times Vacuum electrical breakdown, respectively. It can be seen from figures 6(a) to (e) that as the content of ultramicron tungsten powder in the alloy increases, the erosion pits on the surface of the alloy become significantly shallower. Calibrated and calculated the area of the surface erosion pits. The results are shown in figure 6(f). With the increase of the content of ultramicron tungsten powder in the alloy, the erosion area of the surface becomes larger. That is the addition of ultramicron tungsten powder facilitates the dispersion of the arc erosion, thereby weakening the damage of the arc to the electrical contact materials. At the moment of arc contact, because the lattice structure deforms at the grain boundary, the energy distribution of electrons in the grain boundary and grain is different, and the contact potential difference exists between adjacent grain boundaries and grains. Under the action of external electric field and internal electric field (caused by contact potential difference), the electrons in the grain boundary on
the surface of the breakdown material are more likely to escape from the cathode surface, that is the electron work in the grain boundary is lower than that in the grain. Therefore, materials with more grain boundaries have lower voltage resistance than materials with less grain boundaries. The ultrafine rice flour used in the test not only has lower surface energy, but also less grain boundary energy, so its chemical properties are very stable, which plays a role of dispersing arc at the moment of arc breakdown. Since Cu and W cannot dissolve in each other, the two phases cannot form a solid solution, and due to the poor wettability, the Cu/W two-phase interface is difficult to achieve densification, so its interface bonding is weak, and there are many defects at the interface [13, 14]. Due to the low electron work function and melting point of the Cu phase, during the electrical breakdown of the CuW alloy, the Cu enrichment and the Cu/W phase interface are more likely to be broken down [15, 16]. The particle size of tungsten powder has no effect on the volume and surface area of the Cu phase on the polished surface. While the particle size of tungsten powder has a great influence on the size of Cu/W two-phase interface on the polished surface. When the tungsten powder has a small particle size, the CuW interface is more, but when the tungsten powder is larger, the Cu/W interface is relatively less. The addition of ultramicro tungsten powder is more beneficial to reduce the Cu/W two-phase interface in the CuW alloy, thereby reducing the weak phase structure which is easily broken and increasing the alloy’s withstand voltage strength.

Figure 7 is the 50 times breakdown strength curve of CuW alloys with different tungsten powders grading scheme. From (a) to (e), as the content of ultramicro tungsten powder in the alloy increases, the breakdown strength increases significantly and more and more unstable. Calculated the average value of 50 times breakdown strengths of CuW alloys prepared under different grading schemes to obtain figure 7(f). There are many factors that affect the arc corrosion of alloys, including hardness, melting point, vapor pressure, and electron work function.

According to the theory of thermodynamics and surface chemistry, the chemical potential of block pure material is only a function of temperature and pressure (the pressure has a very small influence on the chemical potential and is usually negligible). And for the highly-dispersed block pure material (powder system), the influence of the surface cannot be ignored. Its chemical potential is not only a function of temperature and pressure, but also relates to the particle size of powder [17].

The larger the particle size of the powder, the lower its chemical potential and higher melting point. Therefore, the ultramicro tungsten powder has the highest melting point, so the content of ultramicro tungsten powder is more, the melting point of the CuW alloy is higher. The melting point of the alloy affects its breakdown strength. In general, the higher the melting point of the alloy, the higher the breakdown strength. Vapor pressure reflects the tendency of a small number of molecules in the matrix to escape from the matrix into the space. The ultramicro W powder has a lower specific surface energy, so the overall energy is also lower, and its ability to sublime into a gaseous state during the breakdown process is weak, so the ability to withstand the...
voltage during the electrical breakdown process is strong. Submicron W powder, on the other hand, has weaker voltage withstand capability. Due to existing contact potential difference, the electrons at surface grain boundaries are more likely to escape from the surface of the material under the internal electric field and the applied electric field \[18\]. Therefore, materials with more grain boundaries have lower voltage resistance than materials with less grain boundaries. The ultramicron powder not only has a lower surface energy, but also has less grain boundaries and a lower grain boundary energy, so its overall electron work function is higher, and the breakdown strength of the alloy is greatly improved.

3.5. Properties of CuW/CrCu monolithic materials

3.5.1. The bonding strength of CuW/CrCu monolithic materials

Figure 8 is the curve about bonding strength of the CuW/CrCu monolithic materials with different tungsten powder grading method. The bonding strength is more than 410 MPa and the maximum is 490 MPa. Xiaohong Yang et al.\[19\] prepared CuW alloy by traditional process and measured its strength to be about 339.04–466.66 Mpa With the increase of the ultramicron tungsten powder (50μm), the bonding strength of CuW/CrCu

![Figure 7](image1.png)

**Figure 7.** The breakdown strength of the CuW alloy with different content of 50μm tungsten powder (a) 15:40:45; (b) 20:40:20; (c) 30:40:30; (d) 40:40:20; (e) 45:40:15.

![Figure 8](image2.png)

**Figure 8.** The bonding strength of CuW/CrCu monolithic materials prepared with different content of 50μm tungsten powder.
monolithic materials is improved. That is, the addition of ultramicron tungsten powder is beneficial to the improvement of the bonding strength of CuW/CrCu monolithic materials.

When the CuW/CrCu monolithic material breaks, the fracture modes of three tungsten powders in the alloy are different. When Yang et al [19] observed the fracture morphology, they found that during the fracture, the fine tungsten particles were torn out and the particles were complete. The dimples formed by the stripping of tungsten particles appeared at the fracture and obvious plastic fracture occurred. However, the larger tungsten particles always break in the middle, showing transgranular fracture. As shown in figure 9, the ultramicron tungsten powder (50μm) undergoes transgranular fracture, a rough fan-shaped pattern is formed on the fracture surface, and the presence of sawtooth can be observed on the fan pattern, which greatly increases the length of the crack and the area of the fracture surface, thus greatly increasing the breaking strength of ultramicron tungsten powder. The micron tungsten powder (6 ∼ 8 μm) undergoes transgranular fracture or intergranular fracture, When the fracture occurs, the crack is extended along the crevice between the grains of the W particle, and formed fracture surface is smooth. The breaking strength of intergranular fracture is smaller due to the small inter atomic strength. The other is the transcrystalline fracture and the fracture strength is relatively high. As shown in figure 9(c), since the tungsten skeleton composed of submicron tungsten powder is discontinuous, the strength of the tungsten skeleton is relatively weak, and the tungsten particles are pulled out of the tungsten skeleton to deselect and produce dimples, resulting in severe deformation and obvious plastic fracture.

The fracture mode of these three kinds of different particle size of tungsten powder determines the fracture strength of ultramicron tungsten powder is the highest, and micron tungsten powder is the second, submicron tungsten powder has the lowest fracture strength. Therefore, as shown in figure 8, the more content of ultramicron tungsten powder, the higher the bonding strength of the CuW/CrCu monolithic materials [20].

3.5.2. Fracture morphology of CuW end

Figure 10 shows the SEM macrographs of the CuW end magnified by 13 times after the different CuW/CrCu monolithic material breaks. It can be seen that when the CuW/CrCu monolithic material breaks, its fracture surface is not smooth with many pits. This rough fracture surface reflects the high bonding strength between CuW/CrCu. It can be seen from the figure that under different grading scheme, the roughness of the fracture surface of the CuW end is different, and the number, size, depth, and dispersion of the pits are all different.

Used software to calibrate the size and number of CuW end pits in the figure 10, the red part is pits as shown in figure 11 below. It can be seen from the calibrated area in the figures that with the ultramicron tungsten powder increase, the regularity of the CuW end depressions is as follows: from less to more, from big to small, from shallow to deep, from centralized to decentralized. The more the pits, the greater the resistance at break. The deeper the pits, the stronger of the structure between atoms on the CuW/CrCu bonding surface at break. The more scattered the pits, the more uniform the CuW/CrCu bonding. The more distributed the pits, the more uniform of the CuW/CrCu bonding surface. The large area of adhesion in figure 11(b) may be caused by tearing. This crack propagation reduces the strength of the bonding surface. Compared with the scattered tearing pits, this large area of tear surface does not have a high breaking strength. Compared with the actual test data of CuW/CrCu bonding strength, the regularity of this change is the same as that of actual data.

Then used software to calibrate the depth of CuW end pits in the figure 10, the calibrated area as shown in figure 12. It can be seen from the calibrated area in the figure that different depths of shades have different grayscale values. Different grayscales are calibrated with different colors, the deepest pits are marked with red, the deeper pits are marked with green, and the shallowest pits are marked with yellow.

From the above figure, we can see that the common features of these five pictures are that the picture of smaller and more uniform pits has more red area, these pits are relatively deeper, and the bonding strength is
higher; while the larger and more continuous pits have more yellow and green area, these pits are relatively shallow, and its bonding strength is also small. Compared these five pictures, the depth of pits on each fracture surface are different in different tungsten powder grading ratios. With the increase of the content of ultramicron tungsten powder, it can be seen that the red marked area on the pit is significantly increased, and the area marked by yellow is obviously reduced. Therefore, with the content of ultramicron tungsten powder increases, the fracture surface of CuW/CrCu has deeper pits, and the bonding strength is higher. This analysis result is consistent with the change regularity of the value in the actual test in figure 8.

4. Conclusions

1. Compared with the conventional CuW alloys, CuW alloys prepared by grading tungsten powder method have good organization and uniformity. The electrical conductivity and hardness are higher more than $20 \sim 30\%$. 

Figure 10. The morphology on CuW surface of CuW/CrCu monolithic materials prepared with different content of 50μm tungsten powder after its fracture. (a) 15:40:45; (b) 20:40:20; (c) 30:40:30; (d) 40:40:20; (e) 45:40:15.

Figure 11. The area calibration on CuW surface of CuW/CrCu monolithic materials prepared with different content of 50μm tungsten powder after its fracture. (a) 15:40:45; (b) 20:40:20; (c) 30:40:30; (d) 40:40:20; (e) 45:40:15.
2. With the increase of the content of ultramicron tungsten powder in the alloy, the erosion craters exhibited shallower and more scattered distribution. The breakdown strength of the CuW alloy also increased from $4.0 \times 10^7$ V m$^{-1}$ to $7.1 \times 10^7$ V m$^{-1}$.

3. The bonding strength of CuW/CrCu prepared by the grading method is higher. With the increase of ultramicron tungsten powder, the bonding strength of CuW/CrCu monolithic materials increased from 410 MPa to 495 MPa. The fracture forms of the ultramicron tungsten powder have a great influence on the bonding strength.

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Figure 12. The depth calibration on CuW surface of CuW/CrCu monolithic materials prepared with different content of 50μm tungsten powder after its fracture. (a) 15:40:45; (b) 20:40:20; (c) 30:40:30; (d) 40:40:20; (e) 45:40:15.
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