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ARTICLE

Effect of Brazing Temperature on Microstructure and Mechanical Properties of Cu-Cr-Zr Alloy

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Abstract: Cu-Cr-Zr alloys are kinds of copper alloys with high strength and high conductivity, usually applied to the manufacturing of key parts in complex environment such as high temperature and wear resistance. In this research, we studied the microstructure, tensile properties, microhardness, and fracture characteristics of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures (600~800 °C) by scanning electron microscope, cupping machine, micro-hardness tester, and stereomicroscope. On this basis, the softening mechanism of Cu-1.04Cr-0.16Zr alloy at high temperature was analyzed. The results show that the ultimate tensile strength (UTS) and microhardness of alloys decrease with the increase of brazing temperature. The UTS, elongation, and microhardness of the samples are about 477.32 MPa, 40.13% and 1517 MPa, respectively. When the brazing temperature of Cu-1.04Cr-0.16Zr alloy is 600 °C, the UTS decreases slightly whereas the microhardness is basically unchanged, and they show good softening resistance. Partial recrystallization starts to occur in the microstructure of Cu-1.04Cr-0.16Zr alloy when the brazing temperature is 650 °C. Some fine and undistorted equiaxed grains appear at the boundary of large equiaxed grains, whose size is 2~7 μm. The pinning effect of precipitates on grain boundary begins to weaken. The UTS and microhardness decrease significantly. The samples are further softened with the increase of brazing temperature, and a lot of annealing twins appear in the microstructure of Cu-1.04Cr-0.16Zr alloy. Cu-1.04Cr-0.16Zr alloy has necking phenomenon and obvious plastic expansion zones after brazing. The shrinkage of cross section increases gradually with the increase of brazing temperature. The dimples grow along the tensile direction, and Cu-1.04Cr-0.16Zr alloy shows good plasticity. In summary, under the condition of satisfying the properties of the brazed joints, the softening of Cu-Cr-Zr alloys can be avoided by brazing filler metals with lower melting temperature.

Key words: brazing; recrystallization; ultimate tensile strength; microhardness

Pure copper is an important functional material of non-ferrous metals, which has excellent conductivity, heat conduction, corrosion resistance, and processing performance, but its application is greatly restricted by its insufficient strength[1-3]. Since the 1960s, a series of high-strength and high-conductivity copper alloys have been developed, such as Cu-Cr-Zr, Cu-Ag, Cu-Mg, Cu-Fe-P, and Cu-Ni-Si[4]. Cu-Cr-Zr alloys are kinds of age-hardenable alloys, whose chemical composition is 0.25 wt%~1.2 wt% Cr, 0.08 wt%~ 0.20 wt% Zr, and balance Cu. They show high strength and hardness, good conductivity, thermal conductivity and wear resistance. Cu-Cr-Zr alloys are important basic and functional materials in modern industry, which are mainly used in the fabrication of International Thermonuclear Experimental Reactor (ITER), integrated circuit, high-power asynchronous traction motor, and high-speed railway[5,6].

The extensive application of high strength and high conductivity Cu-Cr-Zr alloy parts has promoted the development of its welding technology. At present, the welding methods of Cu-Cr-Zr alloys mainly include electron beam welding[7-9], explosion welding[10], friction stir welding[11], diffusion welding[12,13] and brazing[14]. Brazing is a welding method which uses liquid filler metal to fill the gap between the substrate, forming a brazing joint by mutual diffusion

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between the filler metal and the substrate. Brazing has many advantages, such as low heating temperature, small welding stress and deformation. Brazing of Cu-Cr-Zr alloys with the same or different materials has become a key research direction in the world[15]. Li et al[16] achieved the brazing joints of Cu-Cr-Zr barrels and cylindrical holes with the use of Au-based filler metal. The shear strength is higher than 100 MPa. Petrisor et al[17] used Ti-Cu eutectic filler metal to braze Be and Cu-0.6Cr-0.08Zr. The shear strength of brazing joints is above 280 MPa. Singh et al[18] used NiCuMn and TiCuNi filler metals to braze Cu/CuCrZr/SS316L multilayer materials, and the shear strengths of brazing joints are 175 and 150 MPa, respectively. Qu et al[19] adopted Ti-Zr amorphous filler and stress-relieving sandwich material to achieve the brazing joints of W/Cu-Cr-Zr alloy. The shear strength of the joint is greatly improved. Mao et al[20] adopted the brazing joints of graphite/CuCrZr with $CuTiH_2Ni$ filler metal. The resistance of joint area is low, which can meet the application requirements of carbon commutator.

The rapid development of high-speed railway, integrated circuit and other industries has promoted the large-scale application of Cu-Cr-Zr alloys, and the quality for brazed joints of Cu-Cr-Zr alloy is also higher and higher. The brazed joint is composed of two parts: the base material and brazing seam. The reasonable selection and matching of brazing filler metals and brazing processes are the key factors to determine the quality of brazed joints. However, as an important brazing process parameter, the brazing temperature also affects the structure and mechanical properties of the base material in the brazed joints. In this study, the microstructure, tensile strength, fracture morphology and microhardness of age-hardened Cu-Cr-Zr alloy under different brazing temperatures (600~800 °C) were studied, which can provide theoretical reference for the rational selection of brazing filler metals and brazing processes.

1 Experiment

The base material used in this experiment was rod-shaped Cu-Cr-Zr alloy (Φ8 mm×100 mm), of which the chemical composition is 98.8 wt% Cu, 1.04 wt% Cr, and 0.16 wt% Zr. The heating test was carried out in SX2-10-12G resistance furnace. The commonly used brazing temperature was selected as the brazing temperature (600, 650, 700, 750, and 800 °C). The samples were taken out and cooled in the air after holding for 0.5 h. The macroscopic fracture morphology of the samples was observed by Stereo Discovery.v8 stereomicroscope; microstructure and micro fracture morphology were observed by Phenom XL scanning electron microscope (SEM). The formula of microstructural etchant was 10 g $FeCl_3$ with 190 mL distilled water, and the samples were corroded at room temperature for 20 s. The tensile test of Cu-1.04Cr-0.16Zr alloy was referred to GB/T228.1-2010 Tensile Test of Metallic Materials Part 1: Test Method at Room Temperature, and the ultimate tensile strength of samples was tested at a speed of 10 mm/min by MTS C45.105 electronic universal testing machine. The ultimate tensile strength of samples was the average of 5 samples. The hardness of the substrates was analyzed by HV-1000A microhardness meter. At least 5 points were selected for each sample. The loading load was 200 g, and the loading time was 10 s. The arithmetic mean value was taken as the microhardness of the substrate.

2 Results

2.1 Microstructure of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures

Fig.1 shows the microstructure of Cu-1.04Cr-0.16Zr alloy heated at different brazing temperatures for the same time (0.5 h). It is found that there are many coarse equiaxed grains in the microstructure of Cu-1.04Cr-0.16Zr alloy after aging treatment (Fig.1a), of which the grain size and distribution are very uneven. At the same time, there are a few twins in some grains. The migration of grain boundary with a large angle does not occur at 600 °C (Fig.1b), and the grain size and morphology are basically the same as those of the initial state (Fig.1a). With the increase of brazing temperature, Partial recrystallization occurs in Cu-1.04Cr-0.16Zr alloy at 650 °C (Fig.1c), and some fine and undistorted equiaxed grains appear in the large equiaxed boundary, of which the size is 2~7 μm. A large number of annealed twins appear in the grains during the recrystallization process at 750 °C (Fig.1e).

2.2 Tensile properties of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures

Fig.2 shows the tensile stress-strain curves of Cu-1.04Cr-0.16Zr alloy heated at different brazing temperatures. The ultimate tensile strength and yield strength decrease with the increase of brazing temperature, and the elongation increases gradually (Fig.3). Cu-1.04Cr-0.16Zr alloy shows good plasticity as the proportion of uniform plastic deformation stage increases, which gradually exceeds 50% of the deformation. Shown in Fig.3, the initial tensile strength (σ_b) and elongation (δ) of Cu-1.04Cr-0.16Zr alloy are 477.32 MPa and 40.13%, respectively. When the brazing temperature is 600 °C, the ultimate tensile strength of Cu-1.04Cr-0.16Zr alloy is 458.71 MPa, and the elongation is 46.67%. The tensile strength slightly reduces so the Cu-1.04Cr-0.16Zr alloy still maintains a good stability when brazed at 600 °C. According to dislocation theory, the strengthening of precipitated strengthened alloy is mainly due to the interaction between precipitated phase and its strain field and dislocation. Precipitated phase is closely concentrated at the grain boundary, which not only plays a good role in pinning grain boundary, but also hinders the movement of dislocation, and plays an important role in stabilizing the structure[21]. The ultimate tensile strength of Cu-1.04Cr-0.16Zr alloy decreases significantly and the elongation increases substantially at 650 °C or above. The tensile strength of Cu-Cr-Zr alloy is 300.34

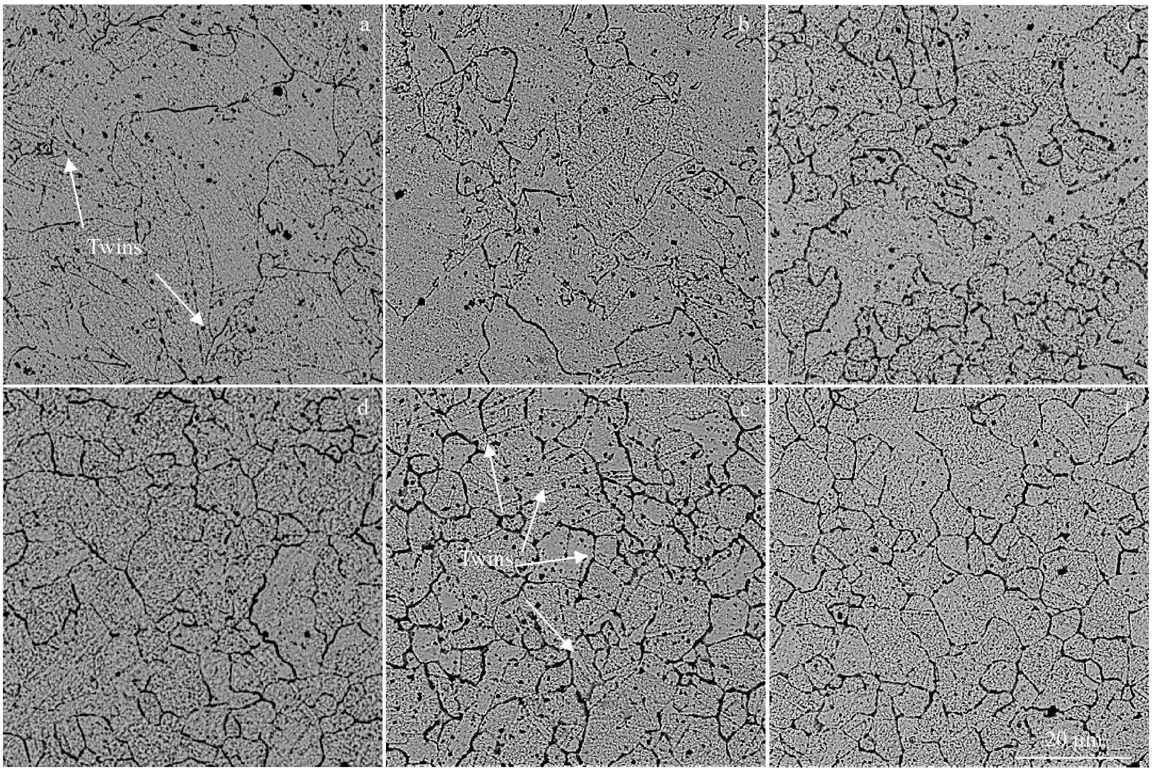

Fig.1 SEM images of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures: (a) original state, (b) 600 °C, (c) 650 °C, (d) 700 °C, (e) 750 °C, and (f) 800 °C

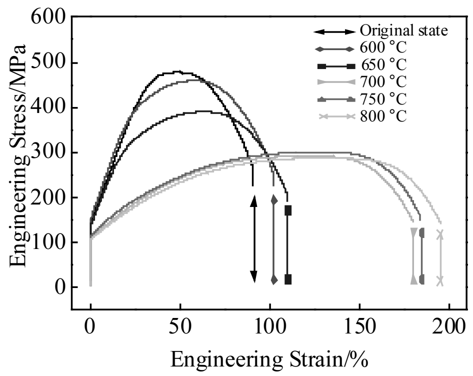

Fig.2 Tensile stress-strain curves of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures

MPa and the elongation is 100.17% at 700 °C, and the tensile strength decreases by about 34.53% compared with the initial state of Cu-1.04Cr-0.16Zr alloy. The rapid decrease of tensile strength is due to the increase of average atomic kinetic energy, the decrease of grain boundary slip and dislocation resistance, the disappearance of pinning effect of precipitate on grain boundary, the nucleation and growth of recrystallized crystal nucleus at grain boundary (Fig.1c), the decrease of dislocation density, and the obvious coarsening of precipitate. The softening effect of recrystallization counteracts the precipitation hardening effect of the phase, which leads to the increase of the plasticity of the alloy and the relaxation of the internal stress. Metal deformation resistance reduces under the action of the tensile stress[21,22]. The ultimate tensile strength of Cu-1.04Cr-0.16Zr alloy decreases slightly and the elongation increases slightly (Fig.3) when the brazing temperature is between 700 °C and 800 °C. The stable recrystallization structure forms (Fig.1d~1f) at those tem-

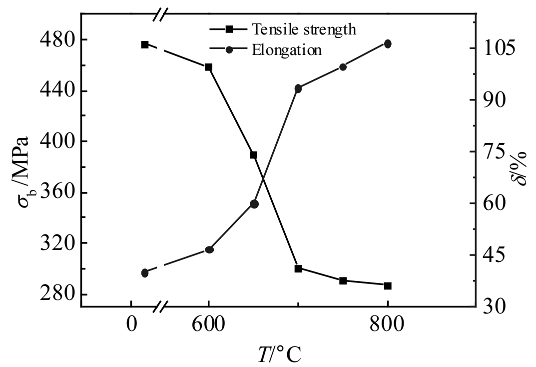

Fig.3 Ultimate tensile strength σ_b and elongation ε of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures

peratures, so the Cu-1.04Cr-0.16Zr alloy shows a gentle softening rate from 700 °C to 800 °C. The tensile strength of the alloy decreases to 286.4 MPa at 800 °C, and the elongation increases to 106.67%.

2.3 Microhardness of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures

The microhardness of Cu-1.04Cr-0.16Zr alloy with the increase of brazing temperature is shown in Fig.4. The initial microhardness of Cu-1.04Cr-0.16Zr alloy is about 1517 MPa. The microhardness of Cu-1.04Cr-0.16Zr alloy is basically unchanged at 600 °C, which suggests that Cu-1.04Cr-0.16Zr alloy exhibits excellent softening resistance when brazed at 600 °C. The microhardness of Cu-1.04Cr-0.16Zr alloy decreases gradually with the increase of brazing temperature when the brazing temperature is 600~800 °C. The microhardness of Cu-1.04Cr-0.16Zr alloy decreases fastest at 600~700 °C. The microhardness of Cu- 1.04Cr-0.16Zr alloy is about 1337 MPa at 700 °C, while when the brazing temperature is 700 °C, the microhardness of Cu-1.04Cr-0.16Zr alloy is about 829 MPa, which is about 54.65% of the initial value of microhardness. After that, the microhardness of Cu-1.04Cr-0.16Zr alloy decreases slowly with the increase of brazing temperature. The microhardness of Cu-1.04Cr- 0.16Zr alloy is 531.9 MPa at 800 °C, which is 64.96% lower than the initial microhardness. The above evidence shows that with the increase of brazing temperature, the plastic deformation ability of Cu-1.04Cr-0.16Zr alloy is getting better and better, and the change trend of microhardness is basically consistent with that of tensile strength.

2.4 Microstructure of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures

Fig.5 shows the macroscopic tensile fracture morphologies of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures. It can be seen that the necking phenomenon occurs in Cu-1.04Cr-0.16Zr alloy at different brazing temperatures, and there is an obvious plastic extension zone, indicating that the fracture mode is ductile fracture. The initial fracture of Cu-1.04Cr-0.16Zr alloy is shown as a tearing fracture with a sectional shrinkage of 69.06%. The fracture is relatively flat and has an obvious “river-like” fold grain, which is perpendicular to the tensile direction. This is caused by the non-uniformity of grain deformation. The plastic expansion area and the shrinkage area become larger with the increase of

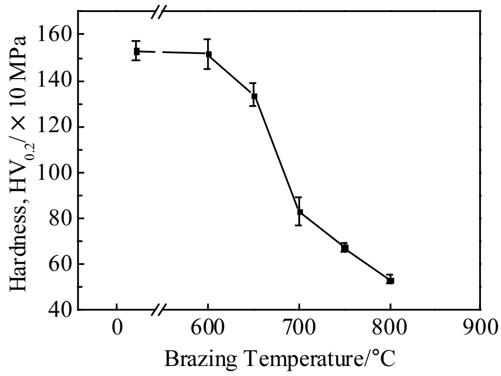

Fig.4 Microhardness of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures

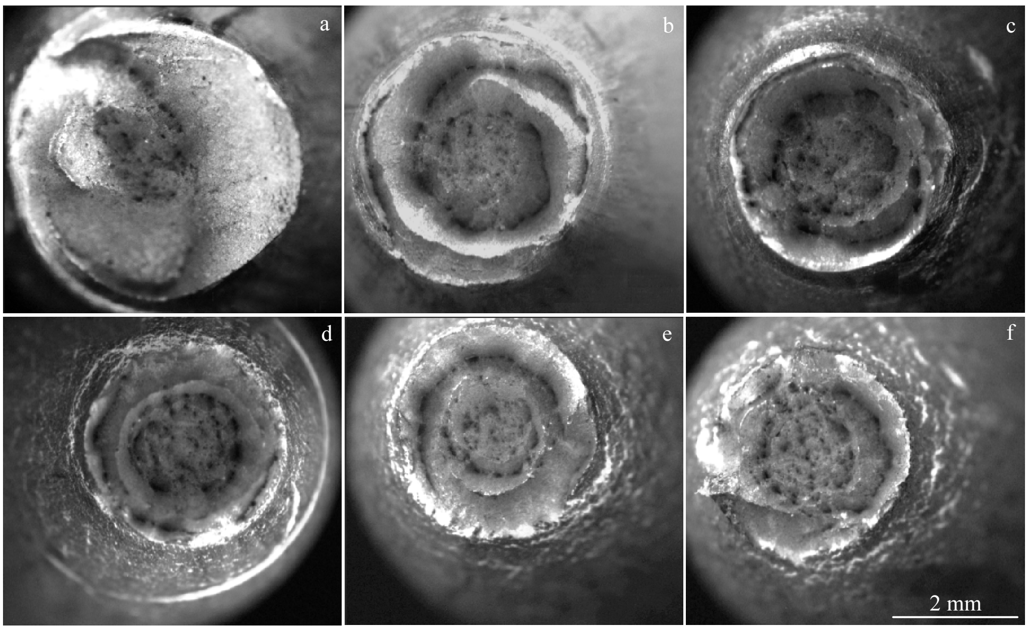

Fig.5 Macroscopic fracture morphologies of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures: (a) original state, (b) 600 °C, (c) 650 °C, (d) 700 °C, (e) 750 °C, and (f) 800 °C

brazing temperature, and the fracture morphology changes into cup-shaped. The shrinkage ratio is 81.42% at 700 °C, and then its increasing trend becomes slower. It suggests that severe plastic deformation occurs in Cu-1.04Cr-0.16Zr alloy before fracture, and the Cu-1.04Cr-0.16Zr alloy has good plasticity at the brazing temperature.

Fig.6 shows the tensile fracture morphologies of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures. It can be seen that the microscopic fracture surfaces of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures are all dimples, and the initial fracture surfaces of Cu-1.04Cr-0.16Zr alloy show the characteristics of tear dimples (Fig.6a). New small dimples appear on the inner wall of dimples with the increase of brazing temperature. The dimples become smaller in size and their density is higher, changing from tearing type to equiaxed type. The reason lies in the recrystallization of Cu-1.04Cr-0.16Zr alloy in brazing process. The recrystallized fine grains interact with the coarsened precipitates under the action of tensile stress. Fracture occurs due to instability, which forms dimples with higher density and smaller size. The dimples of Cu-1.04Cr-0.16Zr alloy are very deep when the brazing temperature is 800 °C, and the stretch marks like beach pattern on the wall of dimples can be observed (Fig.6f), indicating that the alloy has good plasticity at the brazing temperature.

3 Discussion

The softening phenomenon of the material in the heating process is closely related to the change of microstructure. The thermal stability of Cu-Cr-Zr alloy is closely related to the coarsening process of the second phase particles[21]. The coarsening dynamics of the second phase particles can be expressed by Lifshitz-Slyozov-Wagner formula[21,23]:

$$\bar{r}^3 - \bar{r}_0^3 = 8\gamma D C_0 \Omega t / 9RT \quad (1)$$

where $\bar{r}$ is the average particle size at time; $\bar{r}_0$ is the average particle size at the initial stage of coarsening; γ is the interface energy between particle and matrix; D is the diffusion rate of solute atom in matrix; C_0 is the solubility of particle in matrix; Ω is the atomic volume of precipitated phase. When γ, D and C_0 are very small, the coarsening of the second phase particles of Cu-1.04Cr-0.16Zr alloy is very slow, so the Cu-1.04Cr-0.16Zr alloy has excellent thermal stability. In brazing process of Cu-1.04Cr-0.16Zr alloy at high temperature (650 °C or above), D and C_0 increase sharply, so the precipitated phase is easier to coarsen at high temperature. The density of precipitated phase is greatly reduced when the total concentration of solute elements is fixed, and the pinning effect of precipitated phase relative to grain boundary is weakened. The resistance of dislocation movement is reduced, and grain boundary and its strength are reduced. The resistance of grain boundary deformation is reduced, so atoms

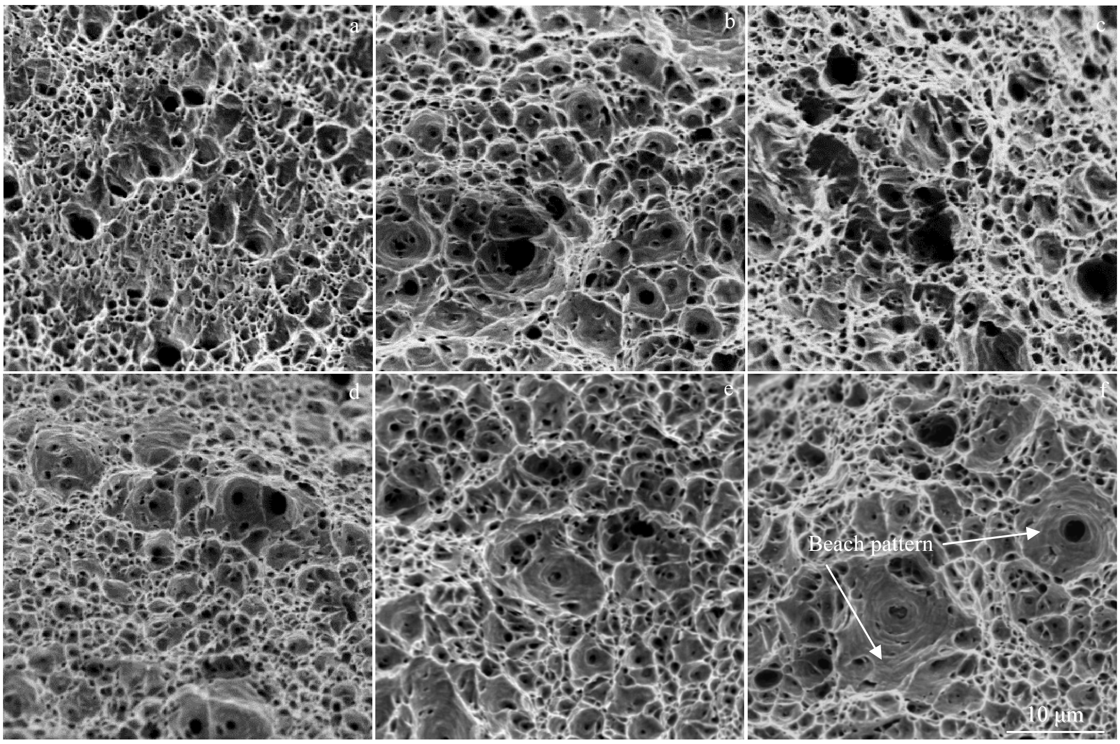

Fig.6 Fracture morphologies of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures: (a) original state, (b) 600 °C, (c) 650 °C, (d) 700 °C, (e) 750 °C, and (f) 800 °C

become easy to disperse. The resistance of grain boundary sliding is reduced, and the grain boundary sliding is sufficient, which leads to the occurrence of alloy softening. Deeper and larger dimples (Fig.6) and some second phase particles are observed.

The thermal stability of Cu-1.04Cr-0.16Zr alloy is also closely related to recrystallization. The precipitated phase and the strain field generated in the aging process hinder the movement of dislocation and inhibit the nucleation of recrystallized grains. At the same time, the migration of grain boundary can be hindered, thus delaying the growth of recrystallized grain. The diffusion rate of solute atoms increases sharply with the increase of brazing temperature, and the precipitated phase gradually coarsens, resulting in a significant decrease in density. Dislocation cells also recover rapidly and become polygonal gradually, resulting in the growth of grain.

The hardness of Cu-1.04Cr-0.16Zr alloy has a mutual restriction relationship with recrystallization fraction. Some researchers believed that the change of hardness can be used to characterize the change of recrystallization fraction[24]. Assuming that the recrystallization fraction corresponding to the hardness H_{max} of Cu-1.04Cr-0.16Zr alloy before brazing is 0, and the recrystallization fraction corresponding to the hardness H_{min} after complete recrystallization is 100%, the hardness H_i at a certain brazing temperature between H_{max} and H_{min} corresponds to a recrystallization fraction X_i, which can be expressed by mathematical expression as follows:

$$X_i = (H_{max} - H_i)/(H_{max} - H_{min}) \quad (2)$$

According to the microhardness results in Fig.4, the recrystallization fraction of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures can be calculated. The hardness values corresponding to 25 and 800 °C are taken as H_{max} and H_{min}, respectively, and the recrystallization fraction corresponding to each brazing temperature is plotted to obtain the recrystallization fraction of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures, as shown in Fig.7. The recrystallization fraction of Cu-1.04Cr-0.16Zr alloy changes only 1.34% at 600 °C, which further proves that the alloy has good stability in this temperature range. When the brazing temperature changes from 600 °C to 800 °C, the recrystallization fraction of Cu-1.04Cr-0.16Zr alloy increases gradually with the increase of brazing temperature. When the brazing temperature ranges from 650 °C to 700 °C, the recrystallization fraction changes rapidly, indicating that the recrystallization is in progress at this stage.

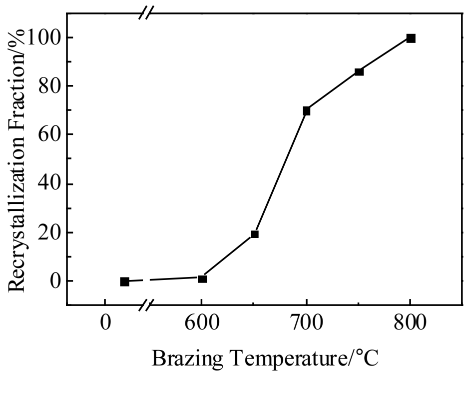

Fig.7 Recrystallization fraction of Cu-1.04Cr-0.16Zr alloy at different brazing temperatures

4 Conclusions

1) The migration of grain boundary with large angle does not occur at 600 °C, and the grain size and morphology are basically the same as those of the initial state. With the increase of brazing temperature, partial recrystallization occurs in Cu-1.04Cr-0.16Zr alloy at 650 °C, and some fine and undistorted equiaxed grains appear in the large equiaxed boundary, of which the grain size is 2~7 μm. A large number of annealed twins appear in the grains during the recrystallization process at 750 °C.

2) The ultimate tensile strength decreases with the increase of temperature, and the elongation increases gradually. The initial tensile strength of Cu-1.04Cr-0.16Zr alloy is 477.32 MPa, and its elongation is 40.13%. The alloy begins to recrystallize when the brazing temperature is 650 °C or above. At the same time, the pinning effect of precipitates on grain boundary disappears gradually, and the tensile strength decreases greatly. The tensile strength of the alloy decreases to 286.4 MPa at 800 °C, and the elongation increases to 106.67%.

3) The microhardness of Cu-1.04Cr-0.16Zr alloy decreases gradually with the increase of brazing temperature. The initial microhardness of the alloy is about 1517 MPa. The microhardness of the alloy remains basically unchanged and shows good anti-softening performance at 600 °C. When the brazing temperature changes from 600 °C to 800 °C, the microhardness of Cu-1.04Cr-0.16Zr alloy decreases sharply with the increase of brazing temperature, and the change trend of microhardness is basically consistent with that of the tensile strength.

4) Cu-1.04Cr-0.16Zr alloy shows obvious necking phenomenon at different brazing temperatures, and there is an obvious plastic extension zone, indicating that the fracture mode is ductile fracture. Dimples extend and grow along the tensile direction with the increase of brazing temperature, and Cu-1.04Cr-0.16Zr alloy shows good plasticity.

5) The tensile strength and hardness of Cu-1.04Cr-0.16Zr alloy decrease sharply when brazed at 650 °C or above. Under the condition of satisfying the properties of the brazed joint, the brazing filler metals with lower melting temperature can be used to avoid the softening phenomenon of Cu-1.04Cr-0.16Zr alloy.

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硬钎焊温度对 Cu-Cr-Zr 合金组织和力学性能的影响

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摘　要：以时效硬化后的 Cu-1.04Cr-0.16Zr 合金为研究对象，采用扫描电镜、拉伸试验机、显微硬度计、体视显微镜研究了不同硬钎焊温度（600～800 ℃）下的 Cu-1.04Cr-0.16Zr 母材的组织、拉伸性能、显微硬度及断口特征，并在此基础上分析了 Cu-1.04Cr-0.16Zr 合金的高温软化机制。结果表明：Cu-1.04Cr-0.16Zr 合金的抗拉强度和显微硬度随钎焊温度的升高而降低，初始抗拉强度为 477.32 MPa，延伸率为 40.13%，显微硬度为 1517 MPa，当钎焊温度为 600 ℃时，Cu-1.04Cr-0.16Zr 合金抗拉强度稍有降低，显微硬度基本不变，表现出良好的抗软化性能；当钎焊温度为 650 ℃时，Cu-1.04Cr-0.16Zr 合金开始发生部分再结晶，大等轴晶边界出现一些细小、无畸变的等轴晶粒，晶粒尺寸为 2~7 μm，析出相对晶界的钉扎作用开始减弱，抗拉强度和显微硬度大幅下降；随着钎焊温度的升高，Cu-1.04Cr-0.16Zr 合金进一步软化，Cu-1.04Cr-0.16Zr 合金内部出现大量的退火孪晶；Cu-1.04Cr-0.16Zr 合金在硬钎焊温度条件下均出现颈缩现象，并存在明显的塑性扩展区，随着钎焊温度的升高，断面收缩率逐渐增大，韧窝沿拉伸方向伸展长大，表现出很好的塑性。在满足钎焊接头各项性能的条件下，可采用熔化温度较低的钎焊材料，避免 Cu-1.04Cr-0.16Zr 合金出现软化现象。

关键词：硬钎焊；再结晶；抗拉强度；显微硬度

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