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Microstructure and properties of liquid film solution-diffusion welding interface for ZCuBe2.5 Alloy①

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Abstract: The microstructures and properties of liquid film solution-diffusion welding interface for ZCuBe2.5 alloy have been studied using Cu-base powder. It reveals that the welding joint has high tensile strength up to 278 MPa, rational distribution of hardness and better matches with base materials in properties. Weld metal consists of the uniform and fine α-Cu equiaxed grain and intergranular $Cu_{5.6}Sn$ phase. The weld is well combined with base materials. The transition solid solution combination interface with a thickness of 150 μm has been formed. In the process of stable welding, the thickness of interface appears to have an increase linearly with bonding time. In the cases of same bonding time, the thickness of interface increases with an increase of temperature gradient, which will become even more apparent with the increase of bonding time.

Key words: liquid film solution-diffusion welding; Cu-based alloy powder; ZCuBe2.5; microstructure; properties
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1 INTRODUCTION

With its many advantages such as powerful malleability, electrical(heat) conductivity, oxidation-resistance and corrosion-resistance, etc, Berkelium bronze plays an important role in manufacturing spare parts of planes and aerospace crafts, etc. At present, the copper alloy connection methods mainly consists of hand arc welding, buried arc welding, gas tungsten arc welding, diffusion bonding and electron beam welding, etc, of which, due to the lower welding temperature and coarsening degree of microstructures, the diffusion welding is easy to obtain high quality welding joints[1-8]. However, the joint structure is confined by the heating and pressure equipments, meanwhile the complicate welding equipment, high cost and long production period also limit its further application. The liquid film solution-diffusion welding (LFSD) is a kind of non-melting base material welding method. It is characterized by diffusion welding and spraying welding[9]. This method can solubilize the base materials on joining interface via the melting of welding materials to form liquid thin film so as to carry out the atom inter-diffusion between welding material liquid film and base materials, whereby the connection with high strength among welding parts can be achieved. In comparison with transient liquid phase diffusion bonding (TLPD)[10-12], this welding method is characterized by simple technological process, high efficiency in welding and strong adaptation to welding structures. Additionally, this method has the advantage of temperature gradient transition liquid phase diffusion bonding(TG-TLPD)[13,14] because of non-iso-temperature feature in the welding process. Based on the analysis of determination of the best spray-melting temperature and its control method[15], the authors study the interface microstructure and properties of steel and iron materials using LFSD welding[9]. This paper deals with the interface microstructures and properties of Berkelium bronze by LFSD welding and provides basis for research and application of solution-diffusion of connecting copper alloy with high properties.

2 EXPERIMENTAL

The U-type grooves with a width of 10 mm and a depth of 10 mm were processed in the middle of rectangular sample with gauge of 13 mm × 13 mm × 130 mm in ZCuBe2.5 alloy. The compositions of the specimen are(mass fraction, %): Co 0.2 - 0.4, Fe < 0.1, Cr< 0.1, Si 0.8 - 1.2, Ni 0.2 - 0.5, Be 2.4 - 2.6, Cu balance. The oxyacetylene flame was used as the thermal resources; and the self-made copper base alloy powders were used as the welding materials. The compositions of the welding material are (mass fraction, %): Fe 14.0 - 16.0, Sn 6.0 - 8.0, P 0.2 - 0.4, a suitable amount of Sb, Cu balance. The size of welding powder is 50 μm. The spray-2/H welding torch was used to carry out the connection of liquid film solution-diffusion on the groove surface in

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the sample. The oxygen pressure and acetylene pressure are 0.7-0.9 MPa and 0.05-0.08 MPa, respectively. The spray melting temperature was controlled according to the liquid film state in the static mirror conditions[15]. The welded samples were processed into bars of d8 mm for mechanical testing; and then, the tensile strength and hardness of samples in the welding zone were measured and tested. The metallographical samples were prepared in terms of standard metallographical technique; and the Newphet-1 optical microscope(OPM) and 4AMRAY-1000B type scanning electron-microscope(SEM) were used to observe the joint structures. Also energy dispersion spectrum (EDS) and D/MAX-1200 type X-ray diffractometer(XRD) were used to analyze the chemical components and phase structures in the micro-zones.

3 RESULTS AND ANALYSES

3.1 Structure characteristics of Cu-base alloy powders

The XRD spectrum of Cu-based alloy powder materials is shown in Fig. 1. It indicates that the solidification microstructures of multi-alloy powder consist of α-Cu and $Cu_{5.6}Sn$ metallic compound phases. Also, owing to the fact that a bulk of Fe and Sn atoms with big radius are solubilized in α-Cu, the diffraction peak slightly moves to the left. Fig. 2 shows the particle morphologies and solidification microstructures of welding powders. In Fig. 2(a), the particle outer shape appears to be irregular oval form, whose ratio of length to thickness is less than (1-4):1; and the smaller size and smooth surface indicate that powders have better fluidity in the process of spray melting. Fig. 2(b) exhibits the powder solidification microstructure, of which light grey is α-Cu phase characterized by uniform and tiny fine equiaxied crystals with grain size of less than 3 μm. The dark grey microstructure located among α-Cu phase is $Cu_{5.6}Sn$ compound which is rich in Sn. Largely owing to the very great cooling rate of powders, the solute trapping occurred in the solidification process. Solid solubility in α-Cu phase increases so that solidification microstructures are in the state of unstable with higher interface energy. The existence of unstable microstructure with higher interface energy can lower the melting point of powder materials so as to improve liquid film fluidity and the interface base material wettability. Therefore, energy input of diffusion connection can be reduced so that the thermal affected zone brittleness and welding part deformation can be avoided and achieve the better connecting joints.

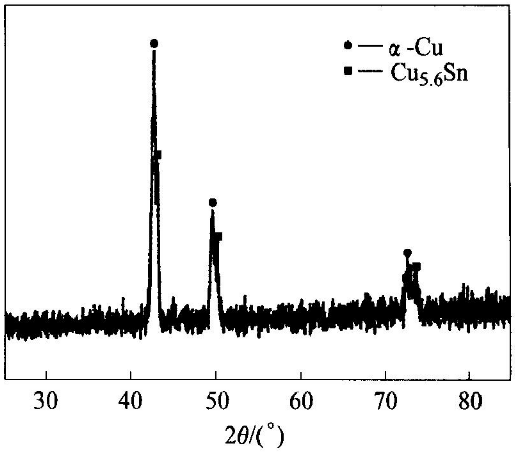

Fig. 1 X-ray diffraction spectrum for Cu-based alloy powder

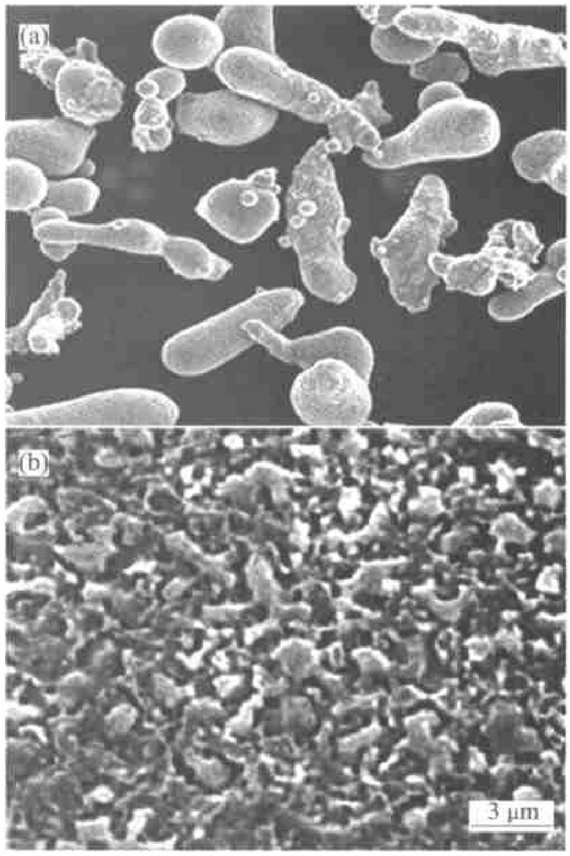

Fig. 2 Morphology and microstructure of Cu-based alloy powder
(a) —Morphology of powder;
(b) —Solidification microstructure

3.2 Microstructure characteristics of connecting joints

Fig. 3 shows the microstructures of liquid film solution-diffusion welding joints. Fig. 3(a) indicates the center microstructure of weld. On the basis of XRD analyses, it is found that after remelting of welding materials and solidification in air cooling, the formed phase structure of weld metals is basically the same as that of powder materials, consisting of α-Cu and $Cu_{5.6}Sn$ phase. However, the Sn-rich phase ap-

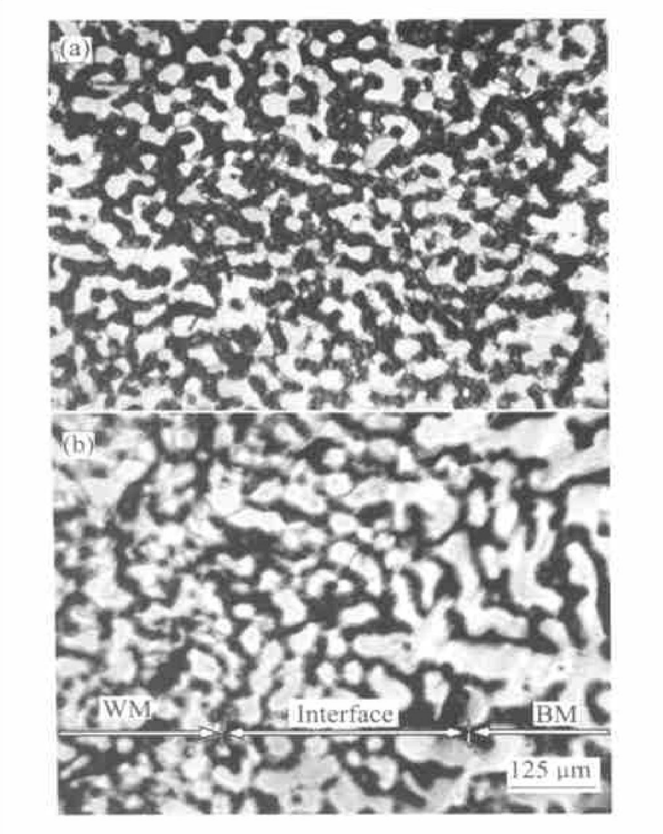

Fig. 3 LFSD joint microstructures for ZCuBe2. 5 alloy
(a) —Weld microstructure;
(b) —Weld/based-metal interface microstructure

parently increases in welding seams. This is related to the fact that weld metal cooling rate is relatively lower than that of powder materials. In addition, as compared to the solidification microstructure of powder materials, the weld microstructure obviously becomes coarsening, but still much finer than that of base materials. The dendritic growth can be seen from α-Cu equiaxed grains in the weld. Therefore, it can be concluded that the initial α-Cu dendrite arm is remelted off due to the release of latent heat, which results in the formation of equiaxed microstructure. What is even more important is that in the process of spray-melting metals layer by layer to form the weld, the liquid film layer under spray-melting has the reheating effect upon the previous sprayed layer in the case of the semi-solidified dendrites, which promotes the dendrites to melt off.

Fig. 3(b) indicates the combining interface microstructure of weld metal and base materials. The better combination of weld metal and base material can form the atom diffusion transition layer with a thickness of about 150 μm. In the diffusion transition layer, weld metal is characterized by the uniform and fine equiaxed crystals and the base metal still maintains the primitive microstructure morphology as shown in Fig. 4, consisting of the coarse α-Cu dendrites and intergranular γ eutectoic phase. The difference is that a part of dendritic arm are melted off and the crystal boundary becomes dull and the size of crystal grains is reduced. Again, the color of interdendrite becomes dark because of being subject to the liquid film solution-diffusion and penetration. In addition, liquid film is very easy to attach and nucleate on the surface of the base materials. Therefore, the epitaxy growth occurs under the greater temperature gradient, and forms the better transition metallurgical combined interface with dense microstructure as shown in Fig. 3(b).

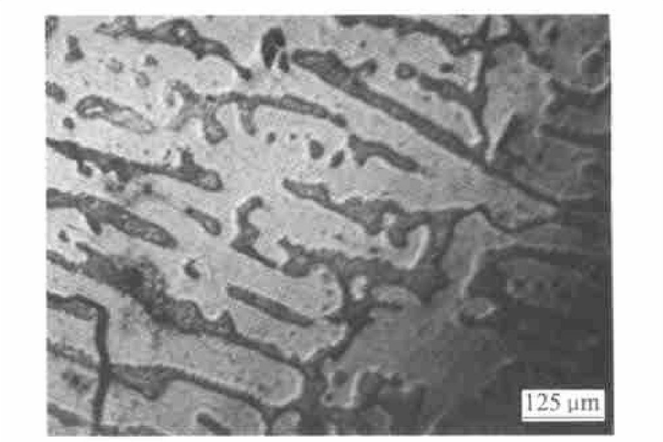

Fig. 4 Microstructure of ZCuBe2. 5 alloy

3.3 Mechanical properties of connecting joints

Atom mutual diffusion via the interface of base materials of liquid film solubilization is carried out, whereby the metallurgical connection with high tensile strength can be obtained. The average value of the joint tensile strength is up to 278 MPa. The hardness distribution of welding joint is shown in Fig. 5. It can be seen that there is a good match between weld metal and base material, but in the combined interface, there appears to have a hardness peak value on the side inclining to base material. This is mainly related to solution intensification and overheating coarsening of microstructure caused by the interface atom inter-diffusion.

4 DISCUSSION

It can be known from Fig. 3(b) that 150 μm-wide diffusion layer formed in the combined interface of weld and base materials because of atom inter-diffusion. Fig. 6 indicates the EDS analyses on the interface. It is easy to see that Cu atoms diffuse from base materials to weld side while Fe and Sn atoms are diffused from the weld to base materials. Fe atom diffusion gradient is large and Cu and Sn atom diffusion

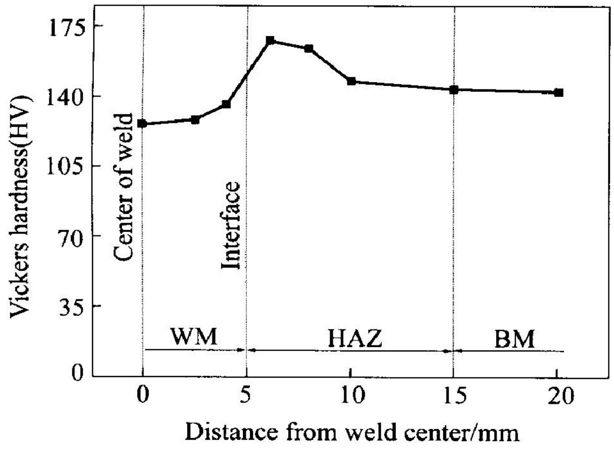

Fig. 5 Distribution of hardness in joint

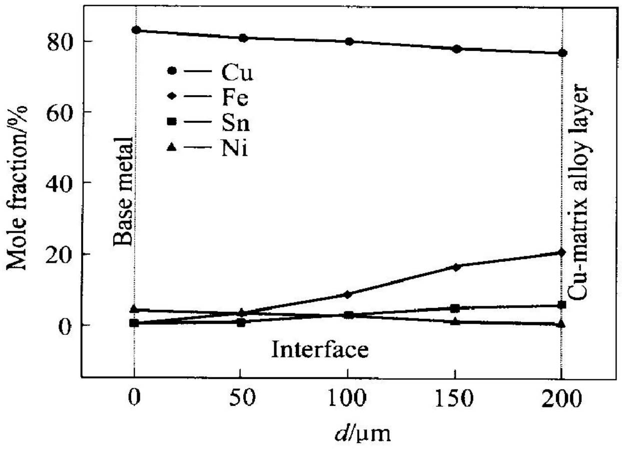

Fig. 6 Mutual diffusion of atoms at interface

gradient is next to that of Fe atom. Since the weld and base materials have lower Al contents, there are no large fluctuation in the concentration in the interface as well as no apparent diffusion in atoms. So, the curve is flat and straight. Since the weld and base metal are all the copper base alloy, and the total amount of alloy element contained in both is not over 25%, in addition, no much difference in Cu, Fe and Sn atom radius, atom replacement performance is so fine and diffusion activating energy is so small that it is easy to realize the instant solution-diffusion connection.

The interface atom diffusion is mainly the atom diffusion in liquid phase[11]. Not only the atom diffusion coefficient is a function of the temperature, but also the interface atom diffusion behaviors are subject to the apparent effect of temperature gradient[13, 14]. Largely owing to technological characteristics of liquid film solution-diffusion welding and geometric symmetry of temperature field along the weld center, the notation method with joint center as the coordinate original point should be adopted in carrying out theoretical analysis. Only considering the single side diffusion, can the feature values of interface diffusion be worked out. Supposing that temperature field does not change with time, and in the solid/liquid interface, with high temperature gradient, the following equation can be obtained using Fick diffusion first law and the principle of interface mass conservation[14]:

$$(C_l - C_0)\frac{dx}{dt} = -D\frac{dC_l}{dx} \tag{1}$$

where C_l is the solute concentration in liquid phase; C_0 is the solute concentration of base material; $D = D_s^{\alpha} \cdot D_l^{(1-\alpha)}$ is the effective diffusion coefficient[11]; D_s is the diffusion coefficient in solid phase; D_l is the diffusion coefficient in liquid phase; α is a constant related to material properties with 0 ~ 0.03 taken in general; $dC_l/dx = (dC_l/dT)(dT/dx) = G/m$ is the solute concentration gradient in liquid phase; G is the temperature gradient in liquid phase; m is the slope rate of liquidus; $C_l = C_{l0} + (G/m)x$ is the solute concentration in the vicinity of interface; C_{l0} is the equilibrium solute concentration of liquid phase in weld center; dx/dt is the migration velocity of characterization interface. When $t = 0$, the interface is located in the half width of weld, i. e., $x = \delta_0$. Substituting it into Eqn. (1), the relation of interface position changing with time can be obtained:

$$\frac{G}{2m}x^2 + (C_{l0} - C_0)x + D\frac{G}{m}t - (C_{l0} - C_0)\delta_0 - \frac{G}{2m}\delta_0^2 = 0 \tag{2}$$

Based on this, the migration velocity, position and diffusion time of diffusion interface of single phase solid solute in double group can be calculated. However, the weld and base materials belong to multi-copper base alloy so that it is difficult to carry out the accurate calculation, but except for high Fe contents, the contents in the rest of alloy elements are low. In order to simplify, Fe diffusion is considered in Cu in calculation. The thermal physical parameters of each element in the above equation are: $D = 3.0 \times 10^{-5}$ cm^2/s, $m = -5^0$C/%, $\delta_0 = 30$ μm, $C_0 = 0.15\%$, $C_{l0} = 15\%$. The relation among the thickness λ of diffusion layer, temperature gradient G, and bonding time t obtained via calculation is shown in Fig. 7. It can be seen from Fig. 7 that the temperature gradient and bonding time in liquid phase can have an apparent effect upon the interface atom interdiffusion. In the process of stable bonding, i. e. in the case of temperature gradient remaining no variation, the thickness of diffusion layer appears to have an approximate linear increase with increasing the bonding time. It can be seen from the different curves in the contrast in Fig. 7 that in the case of the fixed diffusion time the thickness of diffusion layer increases with the increase of temperature gradient. Moreover, the increasing trend will become even more apparent with the increase of bonding time. It has been found that the quantitative results of metallographic analysis are in coincidence with the above

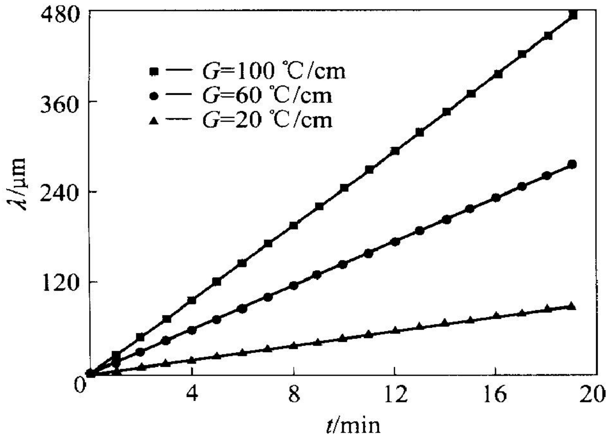

Fig. 7 Interface thickness versus time and temperature gradient

conclusions.

The liquid film solute-diffusion welding is characterized by not only TLP diffusion bonding connection but also the higher temperature gradient than that of TG-TLP diffusion welding, whereby inter-diffusion ability of interface atoms is increased so as to be able to realize the connection with high tensile strength in the shortest possible time.

5 CONCLUSIONS

1) The liquid film solution-diffusion welding of Cu-base alloy powder materials can connect Berkelium bronze. The welding joint has high tensile strength up to 278 MPa. The rational distribution of hardness and better matches with base materials are obtained.

2) Weld metal consists of the uniform and fine α-Cu equiaxed crystal and intercrystal $Cu_{5.6}Sn$ phase. Weld metal is well combined with base materials. The transition solid solution combination interface with a thickness of 150 μm has been formed via the atom inter-diffusion. Interface base materials still remain the microstructure morphology of primitive coarse α-Cu dendrites and intercrystal γ phase, but a part of dendritic arms are melted off and the grain boundary becomes dull and the grain size becomes small to some extent.

3) In the process of stable welding, the thickness of interface appears to have an increase linearly with increasing bonding time. In the cases of same bonding time, the thickness of interface increases with the increase of temperature gradient, which will become even more apparent with the bonding time prolonging.

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