INFILTRATION MECHANISM AND QUALITY CONTROL OF CONTACT MATERIALS①

Wang, Fusheng Zhou, Zaiming Liang, Ronghai
Central-South University of Technology, Changsha 410083, China

ABSTRACT

The infiltration mechanism, which has great significance for the quality control of electrical contact material made from W-Cu, W-Ag alloys with high content of tungsten, has been studied. And a directive infiltration technology for improving the product quality and gaining a better economic benefit has been developed.

Key words: contact material W-Cu alloy W-Ag alloy infiltration mechanism infiltrant leakage black core

1 INTRODUCTION

Electrical contacts made from W_{80}-Cu, W_{70}-Cu and W_{80}-Ag, W_{70}-Ag alloys with high content of tungten have been successfully used for various high tension switches and many electrical equipments due to their superior conductivity, thermal conductivity, wear resistance and fusion-weldability. However the contact materials produced by traditional infiltration technology suffered from "black core"(an un-infiltrated block near the centre of the infiltrafed skeleton), "break-up in spherical shape" (Fig. 1) and "leakage of infiltrant". Those infiltration defects were certainly related to the infiltration mechanism and reduced the qualified grade of their products. Therefore it is very neccessary to study the infiltration mechanism so as to improve the product quality[1].

The quality of the infiltrated products may relate to the distribution state of the refractory skeleton material-solid phase and the fusible infiltrant-liquid phase, and there are generally four states:

(1) The surfaces of the solid phase are not wetted by the liquid phase because of they are not mutually dissolved.

(2) The surfaces of the solid phase are wetted by the liquid phase but the two could not still be mutually dissolved;

(3) The surfaces of the solid phase are wetted by the liquid phase and the former can be partly dissolved in the latter.

(4) The solid and liquid phases could be wetted and mutually dissolved completely.

The infiltration mechanism selected by the authors is basically the same as the second state mentioned above, which means that the molten infiltrant is filled into the porous of the skeleton made from pressed refractory metal powder by capillarity during the infitration. This infiltration mechanism usually has the following advantages due to the small solubility of the refractory solid material.

(1) The deformation of the infiltrated skeleton can be eliminated due to the difference between the melting temperature of the skeleton material and that of the fusible met-

① Manuscript recieved Dec. 20, 1992

(a) (b) (c) (d)

Fig. 1 Micrographs of the sections of infiltrated products

(a), (b) —"black core"; (c), (d)—"break up-in spherical shape"

al is considerably large. For example, the difference between the melting temperatures of tungsten and that of silver is 2 440 C, the difference for tungsten and copper is 2 317 C;

(2)The skeleton material can be wetted well by the fusible metal, and the wetting-anlge θ is always almost small than 90°, which satisfies the following condition fairly well.

$$\text{or} \quad \left.\begin{matrix} (\gamma_S - \gamma_{SL}) > 0 \\ \gamma_L \cos\theta > 0 \end{matrix}\right\} \qquad (1)$$

where γ_S, γ_L and γ_{SL} represemt the surface tensions of the solid phase, the liquid phase and the interface of the solid and liquid phases respectively.

(3)The mutual-solubility of the skeleton material(W) and the infiltrant(Cu, Ag) are either naught or a few if any.

Therefore the technology corresponding to the above mechanism will hopefully be a best one.

2 EXPERIMENTAL

2.1 *Experimental Method*

The experiments were carried out using a specified infiltration technology as follows (see Fig. 2):

Mixing the skeleton material (tungsten powder) with a few additives (nickel powder) and a certain amount of inductive metal (copper or silver) powder→shaping the skeleton via pressing → putting the fusible metal (Cu or Ag) cake beneath the skeleton →infiltrating at a suitable temperature (T) in a molybdenum wire furnace in hydrogen atmosphere→observing the sections of the infiltrated products with a microscope.

Here, adding a certain amount of nickel powder to the skeleton material powder is very neccessary for preforming a W-Ni complex powder in order to improve the wettability of the skeleton material and speed up the infiltration process. And the inductive metal powder is used to form the linking pores prior to the infiltration for speeding up the infiltration process, because this inductive powder can be melted ahead of the infiltration.

2.2 *Infiltration Process*

The infiltration process can be simply described as:

Putting the graphite boat with the

shaped skeleton and the infiltrant cake into a sintering furnace (Fig. 2 (a)) → pushing the graphite boat forward step by step from low temperature zone to high temperature zone, and leading the fusible infiltrant be melted partly (Fig. 2 (b)) and then completely (Fig. 2 (c)) → Infiltrating, a portion of the molten infiltrant is filled upwards into the skeleton pores by capillarity, another portion of the molten infiltrant wets the skeleton sides and meanwhile fill the pores nearby the skeleton sides (Fig2 (d)); and finally all molten infiltrants are infilltrated into the skeleton.

3 ANALYSIS OF THE INFILTRATION MECHANISM

Many factors influence the quality of the infiltration, but only a few are analysed.

3.1 *Capillarity*

Infiltration is essentially a technological process in which the pores of a shaped skeleton are filled with a molten infiltrant by capillary, and mainly used in powder metallurgy for manufacturing compact products. According to Ref. [2], the height (h) risen by the molten infiltrant in a capillary can be expressed as

$$h = 2\sigma\cos\theta / (\rho g r) \qquad (2)$$

where σ — surface tension, N/m;

γ — inner diameter of the capillary;

ρ — density of the molten infiltrant, kg/m^3;

g — gravity acceleration, m/s^2;

θ — wetting angle, (°).

According to Semlak *et al*[3] and assuming the capillaries are vertically parellel each other, the height risen by a molten infiltrant is

$$h = \left(\frac{r\sigma\cos\theta}{2\eta}t\right)^{1/2} \qquad (3)$$

where η — the viscosity of molten infilfrant;

t — infilfration time

Formula (3) indicates that a parabolic relationship existed between h, t and the capillary radius r (Here r corresponds to the radius of the pore in the skeleton), it means that the smaller the particle and the bigger the pore of the skeleton material, the higher the infiltrating height (h) of the liquid phase in a skeleton material will be. However theoretical analysis and practical experience of infiltration showed that there is a limited maximum capillary diameter 0.008 mm and the optimum particle size for infiltration is 4~8 μm.

3.2 *Leakage of Infiltrant*

The phenomenon of a few infiltrant flowing over or leaking out from the infiltrating skeleton is here termed as "leakage of infiltrant", which gives markable influence to the quality of infiltration.

According to formula (3) and supposing

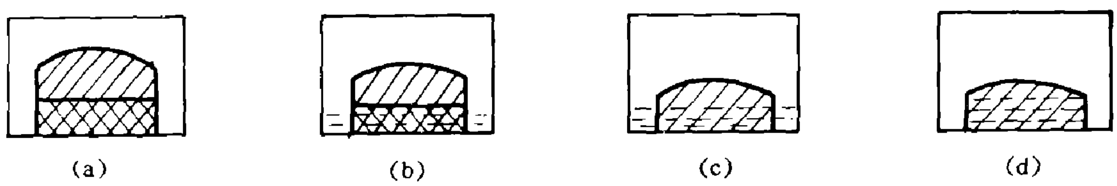

Fig. 2 A schematic diagram of the infiltration process

☐ — tungsten skeleton; ☐ — infiltrant cake; ☐ — molten infiltrant the infiltration temperature (T) to be kept constant(in fact, σ, θ and η are varied with T). the resultant value of σ, θ, η and infiltration time t would also be kept in a certain value P , namely

$$P = (\sigma \cos\theta/\eta)t \qquad (4)$$

Substituting formula (4) into formula (3), we get

$$h^2 = Pr/2 \qquad (5)$$

Formula (5) is a parabolic standard equation, when $r = 0$, $h = 0$, no capillary and infiltration occures; when $r > 0$, h increases with r and P. The larger the P is, the higher the infiltration height will be, and vice versa.

It is worth mentioning again that the P value is influenced by σ , θ and η of the molten infiltrant, and those are linearly decreased with the raising of the infiltration temperafure[1]. However, decreasing P via σ is not beneficial for infiltration, increasing P by η and θ is favourable to infilfration; and when the influence of θ and η become greater than that of σ , the P value will increase. Obviously at a specific temperature, the synthetic influence of θ , η and σ will lead to produce a maximum P value and the corresponding optimum infiltration efficiency, and the specific temperature is correspondingly an optimum infiltration temperature. In addition, different match of particle size in skeleton material and fusible metal has different maximum P value and a correspondingly optimum infiltration temperature, either higher or lower than this optimum infiltration temperature will give rise to decrease the infiltration efficieny, and this is also the right reason for the occurrence of the " leakage of infiltrant".

Based on the above analysis, the authors thought that, measuring the σ , θ and η at a specific infiltration temperature (T) and time (t), and then calculating the maximum and optimum infiltration temperature (T') and using the calculated P , T to direct the infiltration process, the leakage phenomenon could be eliminated.

3.3 *Formation Mechanism of "Black Core"*

(I) Capillary length and infiltration rate

So far some investigations verified that most capillaries located at the side-skeleton are straight line type and those located in the inner-skeleton are essentially curved line type due to the influence of shaping by pressing. For simplification, assuming the capillaries of the inner-skeleton are semicircular links, their average length is now

$$h = \frac{2}{\pi}\left(\frac{R\Sigma \cos\theta}{2\eta}t\right)^{1/2} \qquad (6)$$

Thus, for the inner-skeleton, the capillary length, namely, the infiltration route, is $(\pi/2-1)\,h$ greater than that of the side skeleton. It means the "infiltraion time" of the inner skeleton are longer than that of the side-skeleton.

In addition, capillarity deals with the interface of the solid and the liquid phases, the pre-requisite for the existence of a stable interface during infiltration relates to the generative free energy[5], and the surface tension is often considered as a free energy of a unit area. Therefore the capillarity is closely linked with the surface tension. Different materials have different surface tensions, e.g. for silver (at 1 100 ℃) is 0.879 J/m^2, for copper(at 1 535 ℃) 1.3 J/m^2; yet the same material with the same condifion has the same surface tension. Because the infilfration resistance of the inner skeleton is greater than that of the side skeleton and not vice-versa for the effective surface area, the infiltration rate of the side skeleton is actually greater than those in inner-skeleton.

(2) Influence of infiltration technology

Because the infiltrating skeleton is set on the graphite boat and the infiltration needs time, a portion of the molten infiltrants are flowed upwards via the gaps between the outsides of the skeleton and the inner sides of the boat, and finally cover the top surface of the skeleton and actuate some molten infiltrants to fill the pores nearby the top surface prior to those infiltrants infiltrated upwards.

In general, the infilfration route, resistance and time of the inner-skeleton are normally greater than those of the side-skeleton, and specially a portion of the molten infiltrants reaches the top surface of the skeleton via the gaps between boat and skeleton prior to those infiltrated upwards. As a result, in case of the infiltrant metal cake are set directly under the skeleton, all surfaces and their nearby area are infiltrated before the inner part is infiltrated, and closed certain amount of air in the pores of capillaries of the inner part, which is not filled with molten infiltrant yet. Hereafter, the infiltration becomes more difficult and even forms the "black core"— an un-infiltrated inner block, see Fig. 1(a)～(c)2, 3. From this point of view, the technological method for infiltrating a skeleton, buried completely in infiltrant in hydrogen atmosphere with normal pressure, is not favourable.

3.4 *Cause of "Break-up in Sperical Surface"*

After the inner skeleton has been closed with infiltrated sides, the closed air are compressed and its pressure are raised with the succeeding infilfration by capillarity and the increasing of thermal inflietrtion, which leads to a high radially compressive stress and induced high tangentially tensile stress around the pores with closed air. Consequently a piece or a small layer of infiltrated skeleton be broken-up towards the weakest direction (normally the top surface) under a high stress state, and left a spherical fracture morphology which is coincided with the trajectory line of the tangential principal stress (Fig. 3). Of course, the spherical "break-up" is somewhat influenced by the homogeneity of mixing, the quality of pressing, the pushing velocity of the graphite boat, the rate of infiltration and so on.

3.5 *Directive Infiltration Technology*

Several technologies as partly buried infiltration, buried infiltration and contact infiltration have been used[3], however no one can eliminate the "black core" and the "spherical break-up". The authors thought, based on the above analyses, the "closed air" and the "break-up" could be eliminated provided that the infiltrant is limited to fill the capillary only from one direction. And this

(a)—"break up" has started; (b)—"break-up has displaced; (c)—"break-up" was finished technology is here termed as "directive infiltration".

A simplest method for realizing the idea of "directive infiltration" is that, covering or coating the top surface or/ and other surfaces, in which the infiltration is finished prior to that in the inner skeleton is forbidden or unfavourable.

The covering material should be selected in the light of the wetting-angle (θ) between the solid and the liquid phases. It is realized that $\theta = 170° \sim 180°$ is a preferable selection because of $\theta = 180°$ implies absolutely no mutual-wet between the solid and the liqud phases.

Table 1 Wetting angles and free energy of reaction

Samples	T / ℃	θ /(°)	$F_1 - F_2^*$
Al_2O_3—Sn	1 100	174	−192
Al_2O_3-Cu	1 100	170	−272
MgO-Cu	1 150	160	−101
BeO-V	1 800	35	−49
NiO-Sn	1 200	30	+10
CoO-Sn	900	0	+12

* $F_1 - F_2$ —free energy difference

According to Ref[6], the wetting angle of Al_2O_3-Cu is 170°, see Table 1. Therefore Al_2O_3 powder was selected to cover the top surface of the skeleton, or to bury completely the W-skeleton, with infiltrant cake underneath, in Al_2O_3 powder aimed at avoiding the Cu- infiltrant flows along the gaps between boat and skeleton to the top surface and then infiltrating downwards. The experimental results show that the infiltrated products were free from "black core" and "break-up in spherical shape", the qualified grade of the products increased also from 60 ~ 70% to over than 98%.

4 CONCLUSIONS

(1) A parabolic standard equation induced from Semlak equation can be used to analyze the "leakage of infiltrant", which occurred when the infiltration temperature is higher than an optimum infiltration temperature calculated using maximum P value.

(2) The "black core" is formed when molten infiltrants flow/ infiltrate along the skeleton sides reached the top surface before those infiltrating upwards in the inner part. However the developed "directive infilfration" technology can be used to eliminate the "black core" and the "break-up in spherical shape".

(3) The qualified grade of the infiltrated product has raised from 60~70% to over than 98% by using the parabolic standard equation and the "directive infilfration".

REFFERENCES

1 Tinklepauph, W B; Crandall, W B (ed); Shi, Jin (Trans). Metallic Porcelain. Shanghai: Scientific and Technolgical Press, 1964. 80.

2 Liao, Aixian(ed). Fluid Mechanics. Beijing: Railway Press, 1987. 7.

3 Semlak, K A *et al*. Trans AIME, 1957, 209: 63; 1958, 212: 325.

4 Huang, Peiyun(ed). Principles of Powder Metallurgy. Beijing: Metallurgical Industry Press, 1982. 316.

5 Adamson, A W(ed); Gu, Tiren(Trans). Physical-Chemistry of Surfaces. Beijing: Science Press 1984.

6 Bikerman, J J. Physical Surfaces. New York: Academic Press, 1973. 263.