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Interfacial energy and match of cold pressure welded Ag/Ni and Al/Cu①

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[Abstract] The technology of cold pressure welding was adopted to achieve the bonding of Al/Cu(limited soluble and forming compounds), Ag/Ni (hardly mutual soluble), and the relative state of interface was tested by HREM. The results indicate that stable interface is always corresponding to the low interfacial energy value; the stable interface is not coherent but partly-coherent because of the twisting of grain boundary caused by pressure, meanwhile existing dislocation. Namely, the interfacial match and other states under the condition of cold pressure welding are different from the situation that under the condition of thermal action. Moreover, theoretical analyses and calculation on the base of thermodynamics, crystallogeny, solid physics etc, were discussed.

[Key words] cold pressure welding; interfacial energy; match relationship
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1 INTRODUCTION

The interface refers to the bonding zone of two metals to be welded, and its thickness is only several nanometer(generally several atomic layers)[1, 2]. The interface with peculiarity not only influences the whole joint properties, but also decides its strength, its electroconductivity, and its thermal conductivity etc. It has taken effect on the application of different metals bonding in industry. First of all, the interfacial stability is directly decided by its energy value. For interfacial energy, it may prevent the contracting force when the interfacial surface area is increased.

Assume: E_{ab} —interfacial energy; U_{ab} —unit adhesion work; E_a, E_b —metal surface tension, then:

$$U_{ab} = E_a + E_b - E_{ab}$$

On the view of thermodynamics:

$$E_{ab} = H_{ab} - TS_{ab}$$

where H_{ab} —interface enthalpy; S_{ab} —interface entropy

When there is the lowest interfacial energy, there is the most stable interfacial state. Therefore, the value of energy is always tend to the least during interfacial forming course. The different match of interface will lead to the different interfacial energy, and it is related to the crystal structure, lattice parameter, interfacial formation course etc.

The study of interfacial state can not only reveal the state under the pressure caused by cold pressure welding, but also propel the development and application of bonding technology in solid state, moreover it plays an important role on both theory and practice for development of new materials bonding.

2 EXPERIMENTAL

2.1 Materials

Al, Ag, Cu and Ni with purity of 99.99% were annealed. Their properties are listed in Tables 1 and 2[3~5].

The matches[6] of both Al/Cu (forming compounds) and Ag/Ni(hardly mutually soluble) have great value to give reference to the bonding of different metals.

2.2 Tensile test

The tensile test was carried on a DI-1000B universal tensile testor

1) Al/Ni

Three groups of thread with d1.0 mm were pressure welded, then they were extruded for four times. The average tensile strength was 177.8 MPa.

2) Al/Cu

Table 1 Physical properties of Al, Ag, Cu and Ni

Metal	Atomic mass	Melting point/℃	Density/($kg \cdot m^{-3}$)	Lattice constant/nm	Crystal structure	Heat-transferring ratio/($W \cdot m^{-1} \cdot K^{-1}$)
Al	13	660.24	2.70	4.040	FCC	238.0
Ag	47	961.93	10.50	4.077	FCC	425.0
Cu	29	1 083.00	8.94	3.608	FCC	397.0
Ni	28	1 455.00	8.90	3.516	FCC	88.5

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Table 2 Mechanical properties of Al, Ag, Cu and Ni

Metal	Tensile strength /MPa	Yield strength /MPa	Hardness (HB)	Elongation/ %
Al	80~ 110	50~ 80	15~ 25	32~ 40
Ag	147	55	30	43~ 50
Cu	200~ 240	60~ 70	35	50
Ni	300~ 600	120	90~ 120	10~ 30

Three groups thread with d1.0 mm were cold pressure welded, then they were extruded for four times. The average tensile strength was 275.7 MPa.

For Ag/Ni, Al/Cu, the fracture takes place on the soft metals, which explains the bonding strength of interface is higher than that of the soft metals for the plastic deformation caused by the course of cold pressure welding[7~9].

2.3 Microstructure

2.3.1 Analyses of interfacial components and bonding mechanism

X-ray diffraction method and HREM were used to further study the interfacial state. Al_2Cu deposits on the interface accompanying with the process of bonding under the condition of cold pressure welding (Fig. 1). During the process of cold pressure welding, the chemical attraction appears on the interface of Al/Cu and then the new chemical power presents caused by atomic diffusion. In the end, Al_2Cu deposits on the interface.

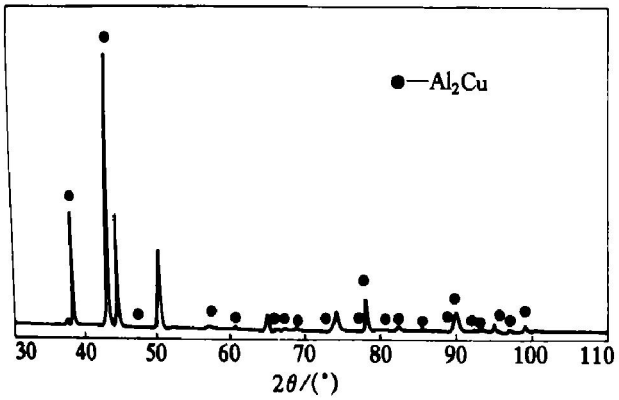

Fig. 1 XRD pattern of Al/Cu interface

No compounds and sosoloid solid solution form on the interface (Fig. 2). During the cold pressure welding, plastic deformation caused many defects and stress etc, which improves the diffusive coefficient and results in the forming of solid solution. This kind of solid solution is not stable, it will deposit with the joint fracturing. Now that the joint has strong tensile strength, its stable bonding results from mechanical bonding and metal atomic bond not from atomic diffusion.

2.3.2 Interfacial match

1) Al/Cu

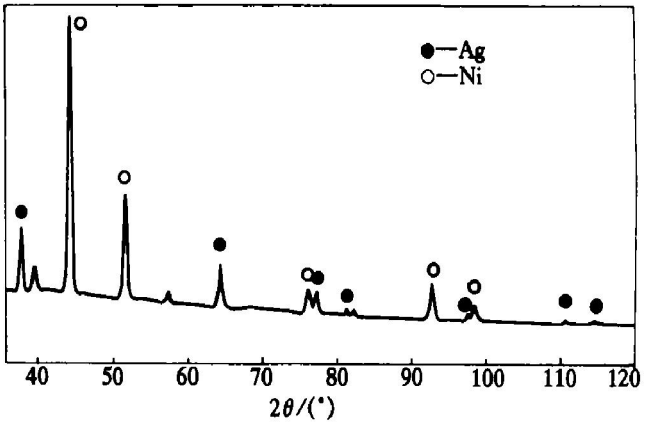

Fig. 2 XRD pattern of Ag/Ni interface

The grain boundary twists under the pressure of cold pressure welding without thermal action, so the partly coherent interface with the match of $(200)_{Al_2Cu} \parallel (111)_{Al}$ forms. On the contrary, under the thermal action, coherent interface with $(110)_{Al_2Cu} \parallel (111)_{Al}$ or $(211)_{Al_2Cu} \parallel (111)_{Al}$ will form.

2) Ag/Ni

Ref. [10] adopted sedimentation method to make Ag/Ni bonding which means that micro-crystal Ag deposit on the base $(111)_{Ni}$, then it is placed on the grid of electronic microscopic telescope to be annealed at the H_2 atmosphere. And the rotation of microscopic crystal observed by TEM do not stop until the state tend to the lower energy. For example, the angle with original 3° then rotating to 44° forms the interface of $(001)_{Ag} \parallel (001)_{Ni}$.

3 THEORETICAL ANALYSES AND CALCULATION

The interfacial match has been one of the important theme in the past decades. People are now especially interested in the interface with low energy that forms along certain dismatch direction.

3.1 Interfacial energy and match of Al/Cu

For metals, semi-coherent or incoherent interface would easily form if there exist different compositions and atomic crystal between the two phase. For approximate compositions, different atomic distance, is usually formed the partly coherent interface, meanwhile, it depends on the dislocation that compensates the different atomic misfit ting. The dislocation is denoted by misfit(δ)[11]:

$$\delta = (\alpha_\beta - \alpha_\alpha)/\alpha_\alpha \quad (1)$$

where α_β, α_α —lattice constant of two phase

On the interface, drastic distortion appearing at the dislocation core, the other atom can match well. In fact, originally, each atom has its most optimal short-range, and its corresponding energy is low.

But, because the changing component on the interface cause the "wrong" atomic bond. On the coherent interface, it only exists the chemical energy. On the incoherent interface, it exists the lattice distortion energy and the chemical energy. For partly coherent interface energy E_{ab}[11] it can be calculated as

$$E_{ab}= Gb/[4\pi(1-\nu)](1-r)^2\{\ln[1/(1-r)] + (1-r/r)\ln(1/1-r)+(r/1-r)\ln(1/r)\}K + r\Delta r \quad (2)$$

Where G—shearing modulus; ν—Poisson ratio; b—Burgers vector model; Δr—the different value of chemical energy between coherent and partly coherent interface; $r\Delta r$—dislocation core energy; $r = m/l$, here m—the length of incoherent interface, $l = b/\delta$ and $0.1 \leqslant r \leqslant 1$; $K = 0.7$.

For Al-Cu: $G = 26$ GPa, $b = 2.863 \times 10^{-10}$ m, $\Delta r = 0.5$ J·m^{-2}, $\delta = 0.12$, $r = 0.7$, then the value of E_{Al-Cu} can be made out as

$$E_{Al-Cu} = 692 \text{ MJ}\cdot\text{m}^{-2}$$

For the interfacial match of Al/Cu, Al_2Cu deposits on interface during the process of cold pressure welding, which is tetragonal system. Its lattice constant $a = 0.6066$ nm, $c = 0.4874$ nm. Under apparently thermal action, the match is $(111)_{Al} \parallel (110)_{Al_2Cu}$ or $(111)_{Al} \parallel (211)_{Al_2Cu}$; but under the cold pressure welding, the match is $(111)_{Al} \parallel (200)_{Al_2Cu}$. the later is very different from the former, and is semi-coherent interface with dislocation.

3.2 Interfacial energy and match of Ag/Ni

The match of Ag/Ni is hardly mutual solube, therefore the theories and experiments about it are seldom reported. Once someone adopted EAM(embed atom method)[12]:

$$F_t = \sum_i F_i(\theta_{h,i}) + \frac{1}{2}\sum_{i,j}\sum_{i,j} V_{i,j}(r_{i,j}) \quad (3)$$

to calculate the E_{Ag-Ni}, the calculation formula is the following, the calculation result is 790~ 895 MJ·m^{-2}.

$$\theta_{h,i} = \sum_j f_j(r_{i,j}) \quad (4)$$

where $F_i(\theta_{h,i})$—the energy that demanded by embedding atom into the location with electron density θ_h; $V_{ij}(r_{i,j})$—interaction potential; $f_{i(r_{i,j})}$—spherical symmetric distribution function of electron density for atom; $r_{i,j}$—atomic distance; $\theta_{h,i}$—electron density at the location i.

For FCC

$$V^{ab}(r) = \frac{1}{2}\left|\frac{f^b(r)}{f^a(r)}V^{aa}(r) + \frac{f^a(r)}{f^b(r)}V^{bb}(r)\right| \quad (5)$$

someone assuming electron density distribution is spherically symmetric and the base electron density is equivalent to the linear superposition of atomic electron density. The two suppositions have deflected the facts. In addition, parameters are decided by the numerical calculation not by the analytic method. The above factors constraint the application of EAM, but EAM suggests a great deal of information for the calculation of interfacial energy of cold pressure welding. Interfacial energy of the different crystal face were tested and are listed in Table 3[10].

From Table 3, we can see that $(111)_{Ag} \parallel (111)_{Ni}$ while $\theta = 0°$, the energy value is 416 MJ·m^{-2}. That is to say the close-packed face (111) not only has the lowest surface energy, but also has the lowest interface energy.

Table 3 Interfacial energy of Ag/Ni(MJ·m^{-2})

Ni crystal face	$(001)_{Ag}$		$(110)_{Ag}$		$(111)_{Ag}$	
	0°	90°	0°	90°	0°	90°
(001)	814	814	1 124	1 124	437	437
(110)	995	995	828	1 214	988	718
(111)	670	670	960	1 008	416	468

This paper further supposes that the crystal lattice is full of the whole space. Generally, they have common points. The lattice location and lattice constant can be adjusted unlimitedly to achieve the four points corresponding coincidence. The four points are not co-face, among them, any three points is not co-line. Being infinititesimal adjusting, the crystal physical nature is not affected. This means that three-location lattices exist, namely, coincidence lattice is caused by the crystal transition symmetricity.

Σ(coincidence density) denotes the ratio of general lattice density and coincidence lattice density.

Theoretically, in order to satisfy the state of low interface energy, the value of Σ should be higher and the value of δ should be lower. But it is impossible that the value of δ is equal to 0 and interface is completely coherent. However low value of δ can be gotten by high value of Σ, or high value of δ can be gotten by low value of Σ(see Table 4).

Table 4 Values of Σ and δ for some crystal faces

Interface	Misfit δ		ΣAg/ΣNi	
	0°	90°	0°	90°
$(111)_{Ag} \parallel (001)_{Ni}$	0.006 0	0.004 3	35/18	8/10
$(111)_{Ag} \parallel (111)_{Ni}$	0.006 0	0.004 3	46/36	4/3

Even the value of δ is very low, but its corresponding interface energy is not the lowest, as listed in Table 3 and 4. For instance, which $\theta = 0°$, $(111)_{Ag} \parallel (001)_{Ni}$, $\delta = 0.004\,3$, the energy is 437 MJ·m^{-2}, not 416 MJ·m^{-2}. Meanwhile, the low value of Σ doesn't means the low energy, for example, $(111)_{Ag} \parallel (111)_{Ni}$, while $\theta = 0°$, ΣAg/ΣNi = 46/36, the energy is 416 MJ·m^{-2}; while $\theta = 90°$,

$\Sigma Ag/\Sigma Ni = 4/3$, the energy is 468 $MJ \cdot m^{-2}$.

From the analysis, in order to achieve the low energy, the value of Σ and δ both need low. Only under the condition, the number of coincidence lattice is larger, the match of crystal lattice is better, the number of fracture bond on the interface is less, so the interface is the most stable one. Consequently, the interface match of Ag/Ni is $(111)_{Ag} \parallel (111)_{Ni}$.

4 CONCLUSIONS

1) The joint strength of Al/Cu is 275.7 MPa; and the strength of Al/Cu is 80~110 MPa larger than that of base Al. The tensile strength of Ag/Ni is 177.8 MPa, for Ag base(being annealed), 147.0 MPa. That is to say the strength of Ag-Ni is higher than that of base Ag. The bonding strength is so strong that it can satisfy the general demands. In addition, for Al/Cu, the bonding mechanism is the results of diffusion action, for Ag/Ni, the bonding mechanism is the results of mechanical bonding and metal atomic bond.

2) For Al/Cu, the partly-coherent interface with the match of $(200)_{Al_2Cu} \parallel (111)_{Al}$ results from grain boundary twisting under the pressure of cold pressure welding without thermal action. On the contrary, under the thermal action, coherent interface with $(110)_{Al_2Cu} \parallel (111)_{Al}$ or $(211)_{Al_2Cu} \parallel (111)_{Al}$ will form. It suggests that the interfacial match of metal joint caused by cold pressure welding is different from the interfacial match of metal joint formed by thermal action.

3) For Ag/Ni, in order to achieve the low interfacial energy, the value of Σ and δ both need low. Only under the condition, the number of coincidence lattice is larger, the match of crystal lattice is better, the number of fracture bond on the interface is less, so interface is the most stable one. Consequently, the interface match of Ag-Ni is $(111)_{Ag} \parallel (111)_{Ni}$.

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