MECHANICAL PROPERTIES OF Si_3N_4/ METAL DIFFUSION WELDING TIP①

Xiong Baiqing
Beijing General Research Institute for Non-ferrous Metals, Beijing 100088
Zhi Rentao, Xiao Jimei
University of Science and Technology Beijing, Beijing 100083

ABSTRACT The mechanical properties of Si_3N_4/ metal diffusion welding tip produced by thermal-vacuum-pressure diffusion welding with pure Al as transition layer were studied. The results show that when temperature, pressure, holding time and cooling rate of diffusion welding are 600 ℃, 80 MPa, 30 min and 2 ℃/ min respectively, the maximum of the shear rupture strength of the tip is 112. 6 MPa. Fracture takes place very easily 1 mm along the Si_3N_4 side of the welding seam. The thermal residual stress comes from the thermal expansion difference between Si_3N_4 and metal. Metallurgical bond forms between Si_3N_4 and metal by interfacial reaction between Al transition layer and Si_3N_4, metal respectively.

Key words mechanical property diffusion welding Si_3N_4/ metal thermal residual stress

1 INTRODUCTION

Si_3N_4 possesses characteristics of high hardness and resistance to oxidation, high temperature, corrosion, and good red hardness, good resistance to thermal shock; so, it is widely used in modern industry. In many practical application, it is necessary to bond Si_3N_4 with metal so as to be used as a whole part[1], besides mechanical bonding, vacuum hard soldering and vacuum-diffusion welding have been studied in many aspects. The former procedure is comparatively simple, but it greatly limits the choice of hard soldering materials owing to the demand of infiltration of Si_3N_4. Vacuum-pressure-diffusion welding has no limits on the choice of materials, but the processing and assembling demands of Si_3N_4/ metal bonding interface are strict, much work has been done on it.

Because the thermal expansion coefficent difference between Si_3N_4 and metal is larger, the residual thermal stress should be considered when Si_3N_4 and metal are bonded together[2]. The residual thermal stress distribution of Si_3N_4/ metal bonding tip is characterized by high stress amplitude and large stress gradient, the finite element method was used to calculate the residual thermal stress distribution of the tip due to lack of precise determining methods[4, 5], but it was affected by many factors, the value and distribution of residual thermal stress obtained always largely deviated from practice. Therefore it couldn't provide reliable basis for quality evoluation of Si_3N_4/ metel tip.

In this case, pure Al was used as transition layer material, and Si_3N_4/ metal welding tip with higher bonding strength was prepared by vacuum thermal diffusion welding. The size and distribution of the residual thermal stress and their effects on bonding strength, rupture location of tip were studied by X-ray micro-area stress analysis.

2 EXPERIMENTAL

2.1 Experimental materials and procedure

The used Si_3N_4 was 3 mm × 4 mm × 8 mm rectanglar pieces after thermal pressing and sintering. The room temperature thermal expansion coefficent α_{RT} is 3.2×10^{-6} ℃$^{-1}$, and the orig-

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inal shear rupture strength is about 400MPa. To compare the effect of matching of thermal expansion coefficent on the residual thermal stress of the tip, higher thermal expansion coefficents of $45^{\#}$ steel ($\alpha_{RT} = 9.1 \times 10^{-6}$ ℃$^{-1}$) and lower thermal expansion coefficent of GH907Fe-Ni alloy ($\alpha_{RT} = 6.37 \times 10^{-6}$ ℃$^{-1}$) were chosen as raw metrials. They were all processed to 8mm × 8mm × 8 mm pieces.

Commercial L_4 pure Al ($\alpha_{RT} = 23.5 \times 10^{-6}$ ℃$^{-1}$) was chosen as transition layer materials. After spark-eroding as pieces, they were ground to 8 mm × 8 mm × 0.4 mm thin slices using $1000^{\#}$ water abrasive paper.

2.2 Vacuum-pressure-diffusion welding

The Si_3N_4 surface for welding were ground by 100 mesh cubic BN powders, then polished. After the surface smoothness of the samples was satisfied, the samples were kept in 6% NaOH boiling water solution for 30 min, then they were put into acetone to be cleaned by ultrasonic wave, and the cleaned samples were conserved in clean acetone.

The $45^{\#}$ steel and GH907Fe-Ni alloy samples were ground by water abrasive paper till $1000^{\#}$, then polished. After the smoothness of the samples was also satisfied, they were put into acetone to be cleaned by ultrasonic wave, then were conserved in clean acetone to aviod surface oxidation in air. The equipment used was a small vacuum pressure diffusion welding machine whose vacuum is no less than 9.31×10^{-5} Pa. The samples were piled up in vacuum chamber from bottom to top in the order of metal, transition layer and Si_3N_4, pressed at 80 MPa, vacuumed to 6.55×10^{-3}Pa, then heated, the heating rate was controlled within 10 ℃/ min till temperature was up to 600 ℃. After the samples were held for 30 min at 600 ℃ and the cooling system began to work, the cooling rate was controlled within 2 ℃/ min till room temperature by modifying the cooling water flow rate continuously, then vacuum and pressure were disannualled and samples could be taken to be tested.

2.3 Strength test of tip

The tip samples were grasped by special jig, the tip surfaces suffered shear stress, and were tested on a mechanical test machine at a rate of 1.0 mm/ min till they ruptured. The maximum of shear stress was calculated by taking the real welding area α as the calculating area α, and it was used to stand for the strength of tip.

2.4 X-ray diffraction micro-area stress analysis

A Japnese MSF-2M X-ray stress analysis instrument was used. The samples tested were covered partly, i. e. a Pb plate with a slot was fixed over the samples, the diffusion interface of samples was adjusted to the slot of Pb plate. The micromoving of samples was carried out on a special sample stand (Fig. 1) . Therefore, the residual thermal stress of locations whose distances to the diffusion welding interface were different was determined.

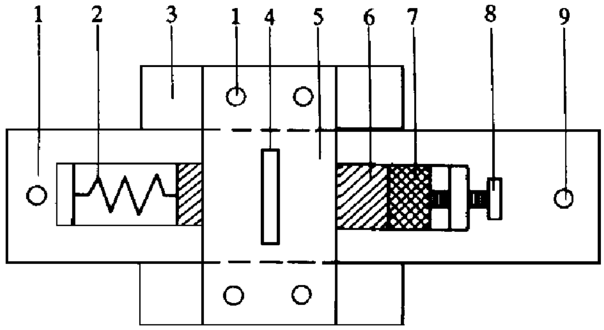

Fig. 1 Schematic diagram of sample stand for X-ray diffraction micro-area stress analysis

1 —sample stand; 2 —spring; 3 —Pb plate stand; 4 —slot; 5 —Pb plate; 6 —sample; 7 —driving head; 8 —lead screw bolt; 9 —fixed screw bolt

In shear experiment, the rupture took place at the side of Si_3N_4, which indicated where the weakest part of the tip was. Therefore, in the X-ray diffraction micro-area analysis, the residual thermal stress distribution of Si_3N_4 side near welding interface was emphatically studied by the $\sin^2 \Psi$ method being taken.

2.5 Gradient and phase analysis near diffusion welding interface

The morphology near the diffusion welding interface of the tip was observed using a Japanese JSM-35CF SEM, and the element distribution was analyzed using its attached wave spectrum equipment by wave spectrum face scaning method.

The experimental samples were thinned from metal side stratum by stratum, and the X-ray diffraction was conducted in a Holland Phillips APD-10 diffractometer after each thinning.

3 RESULTS

3.1 Strength and rupture location of tip

In the shear rupture test of Si_3N_4/metal tip, all the ruptures took place 1 mm from Si_3N_4 side of the welding interface. The average value of shear strength of Si_3N_4/GH907 alloy tip was 112.6 MPa, that of Si_3N_4/45# steel was 92.3 MPa.

3.2 Distribution of residual thermal stress of tip

In general cases, the main reason which caused the tip to rupture at the side of Si_3N_4 is the normal stress σ_y perpendicular to the direction of welding surface. The relationship of σ_y with x from the Si_3N_4 side of the tip obtained by X-ray micro-area diffraction analysis is shown in Fig. 2.

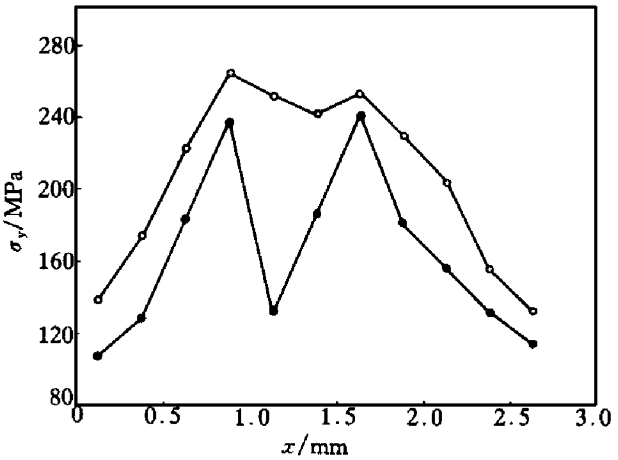

Fig. 2 Distribution of σ_y on Si_3N_4 side of tip
○—45# Steel; ●—GH907

3.3 X-ray diffraction phase analysis

The results are shown in Fig. 3 and Fig. 4.

3.4 SEM observation

A scanning electron microscope was used to observe the morphology of Si_3N_4/metal interface. It is found that in Fig. 5 the interface bonding is good and there are no obvious cracks.

The wave spectrum element face scanning analysis method was used to analyze element distribution of Si_3N_4/metal tip. It is found that there exist much Si, Fe, Ni elements in Al transition layer which indicates the elements abovementioned obviously diffused towards interior of Al transition layer.

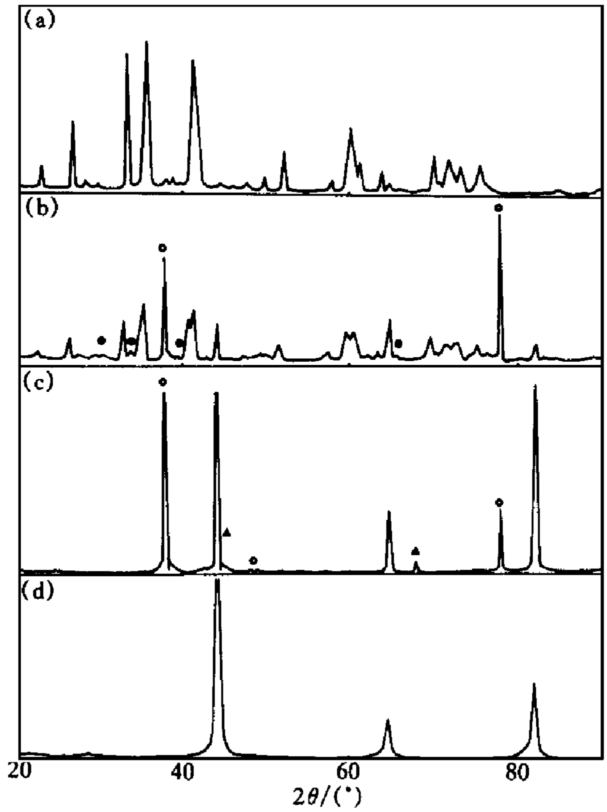

Fig. 3 X-ray diffraction spectra of different locations of Si_3N_4/45# steel tip
(a) —Si_3N_4; (b) —Al/Si_3N_4 interface;
(c) —45# alloy/Al interface;
(d) —45# steel
● —AlN; ○—Fe_2Al_5; ▲ —Fe_3Al_5

4 DISCUSSION

4.1 Bonding mechanism of tip

It can be seen in the X-ray diffraction spectra (Fig. 3(b), Fig. 4(b)) that the AlN peak appears and then it may be deduced that the following reaction takes place:

$$4Al + Si_3N_4 \longrightarrow 4AlN + 3Si.$$

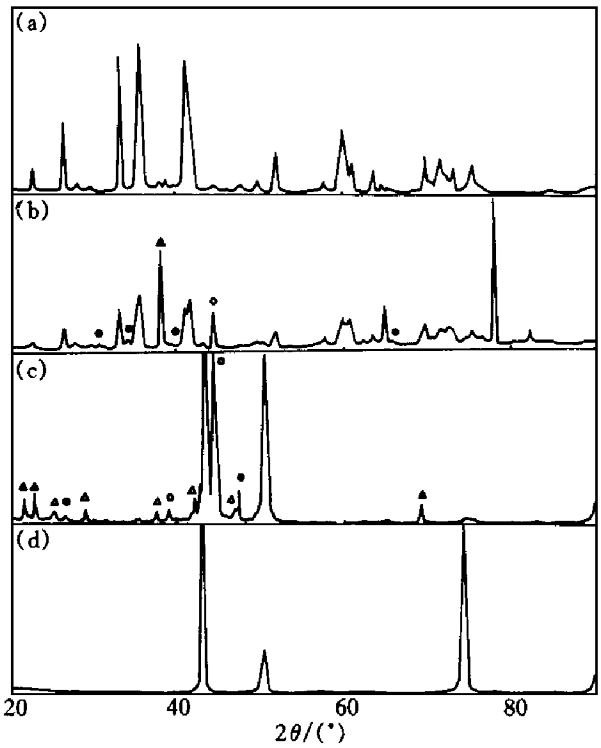

Fig. 4 X-ray diffraction spectra of different locations of Si_3N_4/ GH907 alloy tip

(a) —Si_3N_4; (b) —Al/ Si_3N_4 interface;
(c) —GH907 alloy/ Al interface;
(d) —GH907alloy
● —AlN; ○—Fe_2Al_5; ▲ —Fe_3Al; △ —Ni_3Al

It is known from thermodynamics that when diffusion temperature is 600 ℃, the ΔG of the reaction above-mentioned is − 112. 5 kJ/ mol (< 0), therefore the reaction can take place in this experiment. Besides, it can be seen in spectra of Fig. 3 (c) and Fig. 4 (c) that, Fe_3Al, Fe_2Al_5, Ni_3Al peaks appear. With the aid of related thermodynamics handbooks, it is verfied that the interfacial reaction can take place between Al and 45# steel or GH907-Ni alloy in this experiment. It is reasonable that the formation of Si_3N_4/ metal tip mainly depends on interfacial reaction between Al transition layer and Si_3N_4, Al transition layer and metal, and the firm metallurgical bonding forms.

4. 2 Rupture form of tip

Generally speaking, in this experiment, the location of Si_3N_4/ metal tip where rupture took place most easily should be the interior of pure Al transition layer, because the strength of pure Al is lower not only than those of 45# steel and GH907Fi-Ni alloy but also than that of original Si_3N_4. From the results of shear rupture strength of the tip, the rupture of Si_3N_4/ metal diffusion welding tip took place at the Si_3N_4 side, meanwhile the shear rupture strength value of tip is higher than that of pure Al, but lower than that of original Si_3N_4. The decrease of the strength of the Si_3N_4 side of the tip must be related with the residual thermal stress caused by the thermal expansion coefficent difference between Si_3N_4 and metal. The increase of the strength of pure Al indicates pure Al transition layer was strengthened in someway by reaction diffusion in thermal pressure diffusion welding.

From the results of SEM analyses, there existed obvious long-distance diffusion of Fe, Ni from the metal side towards the pure Al transition layer in the tip. From the X-ray diffraction spectra, it can be known that the diffusion resulting in the reactions between Al and Fe, Al and Ni, formed intermetallic compounds such as

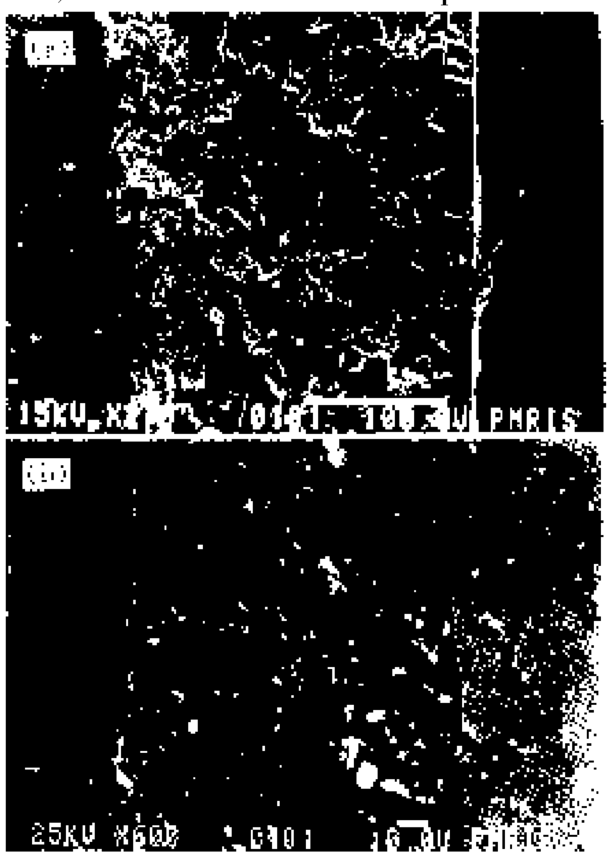

Fig. 5 Morphologies of Si_3N_4/ metal diffusion welding interface

(a) —45# steel/ Al/ Si_3N_4;
(b) – Si_3N_4/ Al/ GH907 alloy

Fe_3Al, Fe_2Al_5 and Ni_3Al. If the intermetallic compound particles dispersively distributed in the Al transition layer, the Al transition layer was certainly be strengthened; besides, Si has higher solution in Al, when Si resulting from the reaction between Al and Si_3N_4 diffused into the Al transition layer and formed solution solid, the Al transition layer was strengthened.

Summing up the analyses, it can be deduced that the Al transition layer strength mainly depends on the following two factors: one is dispersive strength of the intermetallic compound particles produced by the reaction between Al and Fe, Al and Ni; the other is solid solution strength of Si resulting from the reaction between Al and Si_3N_4.

4.3 Effect of residual thermal stress

Before the formation of diffusion welding tip, the shear strength of Si_3N_4 is about 400 MPa, but after the formation of the diffusion welding tip , the shear rupture strength of Si_3N_4 side of the tip is only about 100 MPa, that is even lower than that of Al transition layer strengthened. The reason is that the thermal exepansion coefficent difference between Si_3N_4 and metal is larger. When the tip cooling down, two sides of interface will certainly result in residual thermal stress, but Si_3N_4 is fragile material, its plastic deformation ability is poor, its stress cannot be released, which will result in stress concentration at the side of the Si_3N_4 after the tip cooling down. It can be seen from Fig. 2 that there exists very large residual thermal tensile stress at the Si_3N_4 side of tip, for Si_3N_4 its tensile strength is far lower than its compression strength, therefore, when interior residual thermal stress of Si_3N_4 is tensile stress and its value is larger, it certainly results in self-strength obvious decreasing. When the strength decrease is up to a certain value which is lower than that of other parts of the tip, rupture takes place primarily at the Si_3N_4 side of the tip. It can also be seen from Fig. 2, there is a residual thermal tensile stress peak at the Si_3N_4 side where was 0.8 ~ 1.0 mm from the welding interface. Compared with the shear rupture test results, it is found that the rupture just takes place near the site, which explains the effect of residual thermal stress on the rupture form of the Si_3N_4/metal tip.

It is known from Fig. 2 that the residual thermal stress of every part of Si_3N_4/GH907 alloy tip is much lower than that of Si_3N_4/45$^{\#}$ steel tip, but the shear strength of the former is higher than that of the latter, it indicates that when the metal whose thermal expansion coefficent is near that of Si_3N_4 is chosen to weld with Si_3N_4, the residual thermal stress could decrease and the strength will increase.

5 CONCLUSIONS

(1) In diffusion welding, dispersive strengthening resulting from Fe, Ni of metal side diffusing towards Al transition layer and solution strengthening resulting from Si from the Al/Si_3N_4 interfacial reaction diffusion towards Al transition layer, are two significant factors contributing to Al transition layer strengthening.

(2) When diffusion temperature, pressure, holding time, rate of cooling are 600 ℃, 80 MPa, 30 min, 2 ℃/min, respectively, the maximum values of shear strength of Si_3N_4/45$^{\#}$ steel tip and Si_3N_4/GH907 Fe-Ni alloy tip are 923 MPa and 112.6 MPa, respectively, and rupture takes place at Si_3N_4 side —1 mm from welding interface. whose residual thermal stress is the largest.

(3) The residual thermal stress comes from thermal expansion difference between Si_3N_4 and metal.

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