

The Brazing Behavior of Aluminum-Steel Cladding Strip with Pre-Cladded Brazing Layer Used in Air Cooling System of Power Plants

Chang SUN^{1,a}, Xiao-jun ZHANG², Long LI³, De-jing ZHOU⁴

^{1,2,3,4}Yin Bang Clad Material Co., Ltd., Wuxi Jiangsu, China

Abstract. The brazing behavior of 3003 fin assemble with 4343 (filler layer)/4A60 (transition layer)/08Al (steel layer) brazing sheet (the ratio of filler layer thickness to transition thickness were constant 1:2) were investigated. This study elucidates the brazing results vary with the layer thickness and brazing temperatures. Results showed that keeping the holding time at brazing temperature constant (the holding time of this study was 10 minutes in all brazing progress), under the condition of brazing temperature less than or equal to 580 °C, thinner filler layer led to well formed joint, while thicker filler layer led to defects at brazing joints because of poor mobility. When brazing temperature was up to 600 °C, a thinner transition layer trended to lead to the formation a thick intermetallic compounds layer on the Aluminum-Steel surface. It was inferred that Si diffused from molten filler metal to the interface of transition layer and steel layer and promoted the formation of intermetallic compounds. Furthermore, it was found that in the surface away from brazing joints, with lower Si content, the intermetallic compounds were mainly η (Fe_2Al_5), while higher Si content region nearby brazing joints trended to form τ_5 ($\text{Al}_8\text{Fe}_2\text{Si}$) and τ_6 ($\text{Al}_{4.5}\text{FeSi}$). Meanwhile, a transition layer with thickness above 134 μm could avoid the formation of intermetallic compound. The optimum brazing process was that temperature range was 590 °C to 595 °C with 10-minutes holding time and the transition layer thickness was suggested to be equal or more than 134 μm .

1 Introduction

With the development of electric power technology, the high parameter steam turbine unit of the thermal power plants are increasing year by year. So there is a greater demand for water resources in the cooling system of the power plant. Air-cooling system for power

* Corresponding author: dejing.zhou@cn-yinbang.com

plants was developed in 1970s in order to alleviate the contradiction between the shortage of water resource and the increasing water demand in power stations. Not only can the air-cooling system improve the efficiency of heat exchanger, but it also can save at least 75% of the water compared with the traditional ways [1].

One of the key technologies of producing air-cooling system for power plant is the manufacturing technology of the tube-fin heat exchanger. The air-cooling tube-fin exchanger is mainly manufactured by brazing the base tube and the fin. Base tube, made of aluminum-steel clad prepared by rolling-bonding process, has both good mechanical properties of the steel and good thermal conductivity of aluminum alloy[2-3]. The fin, made of aluminum alloy which has good thermal conductivity, is brazed on the base tube. Heating up to melt the brazing filler, with the aid of capillary force, the filler filled into the gap between fins and base tube so that brazed joint formed by metallurgical bonding after solidification[4]. Aluminum-steel cladding strip with pre-cladded brazing layer has been developed in recent years. The filler layer is hot rolled with transition aluminum layer. The as-rolled sheet and steel are cold rolled by the rolling mill. Then, the monolayer layer fin and the three layer aluminum-steel clad are brazed. This can reduce the times of rolling and save cost. Besides, the filler coating on the surface of aluminum-steel clad is benefit to avoid erosion of the fin.

The brazing temperature and the thickness of transition layer are the key parameters of brazing process. During this process, it is prone to cause erosion defects of the transition layer. After the transition layer was penetrated by the molten filler, the Si element accumulated at the interface of aluminum-steel, which leads to the formation of brittle compounds at the interface of aluminum-steel. The brittle compounds can severely damage the bonding of the aluminum and steel, weaken the mechanical properties of material[5]. Thus it greatly reduced the service life of the material. In this paper, the monolayer 3003 fin and three layers of aluminum-steel clad material were brazed at different temperature and different thickness of the 4A60 transition layer by controlled atmosphere brazing(CAB) process. Silicon's effect on the erosion defects of the transition and formation of the intermetallic compound(IMC) at the interface was investigated.

2 Experimental Procedure

Three layers of aluminum-steel cladding strip 4343/4A60/08Al (filler layer/ transition layer/steel) with different thickness and 3003 fin were chosen as the experimental materials. Elements and specifications of the materials are shown in table 1 and table 2.

Table 1 elements (wt.%) and properties of the fin and three layers of aluminum-steel cladding strip

Elements	Si	Fe	Cu	Mn	Zn	Ti	Al	solidus	liquidus
3003(fin)	0.55	0.5	0.125	1.15	-	0.02	bal	643 °C	654 °C
4343(filler layer)	7.5	0.19	0.01	0.03	0.005	0.01	bal	577 °C	613 °C
4A60(transition layer)	0.81	0.28	-	-	0.01	0.02	bal	642 °C	655 °C
08Al(steel)	-	bal	-	0.25	-	-	-	>1000 °C	>1000 °C

Table 2 thickness of the layers of aluminum-steel cladding strip and aluminum layer

(filler layer:transition layer \approx 1:2)

Thickness(mm)	Filler layer(μ m)	Transition layer(μ m)
2.03	62	112
2.47	76	134
2.70	87	155

Firstly, the specimens were scrubbed with alcohol to remove grease and other impurities on the surface. And then the specimens were soaked in the NaOH solution with mass fraction of 10% for 2min, so that the oxide film on the the specimens was removed. On the filler surface of the aluminum-steel cladding strip, fluoaluminate flux suspension (NOCOLOK, AlF₃-KF) with mass fraction of 10% was painted after rinsed. Then the specimens was dried at 80 °C in the oven. The fin was placed on the filler surface of the aluminum-steel cladding strip and ensure good contact between the two. Then the assembles were put into the brazing furnace to carry out brazing under the condition of Ar gas protection.

During the brazing process, to make the specimens heated uniformly, take the stage of heating and thermal insulation. The furnace was first evacuated to a vasumm less than 100Pa, purged with high-purify (99.999%) Ar, and then heated to 500 °C at the rate of 20 °C /min in the flowing Ar. The furnace was heated to 550 °C at the rate of 5 °C /min after holding at 500 °C for 10min. And then, the furnace was heated to brazing temperature in 10min after holding at 550 °C for 10min. After holden at brazing temperature for 10min, the furnace was cooled to room temperature at a rate of 30°C/min. Brazing temperature was 570 °C, 575 °C, 580 °C, 585 °C, 590 °C, 595 °C, 600 °C, respectively. In this experiment, all holding time at final brazing temperature were 10 minutes.

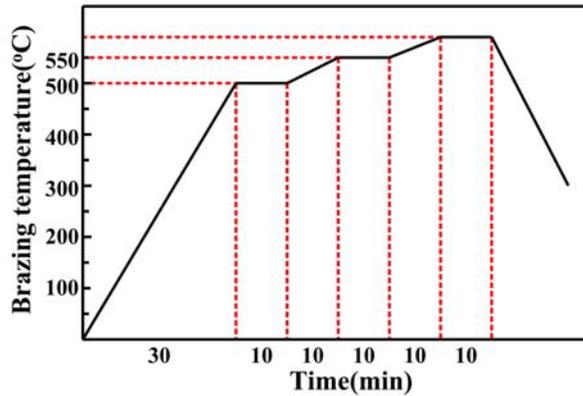


Fig.1 Temperature rising curve of brazing process

The interface morphology between the transition and steel layer was observed by Zeiss optical microscope. JSM scanning electron microscope (SEM) and EDAX energy dispersive spectrometer were utilized to study the microstructure and various phases of the interface.

3 Results and Discussion

3.1 Effect of Temperature on Brazing Behavior

Figure 2 and table 3 shows the different experimental results after brazing at different temperature. Brazing temperature can affect the liquidity of the filler effectively. It was able to form brazed joint when the thickness of the filler layer was $62\mu\text{m}$, under the condition of $570\text{ }^{\circ}\text{C} \sim 580\text{ }^{\circ}\text{C}$. However, in figure 3a-b, there were parts of the position can't form metallurgical combination. But when the filler layer thickness $\geq 76\mu\text{m}$, the filler cannot be fully melted. Owing to poor liquidity of the molten filler, the capillary force cannot completely overcome viscous resistance force. Therefore in figure 3f-g, only a small part of the filler flowed to the brazed seam, formed brazed joint with defects. Figure 3c-d and h-j show that when the brazing temperature range was $590\text{ }^{\circ}\text{C} \sim 595\text{ }^{\circ}\text{C}$, the filler melted with good liquidity. The capillary force can overcome viscous resistance force. The filler flowed to the brazed joint, formed perfect brazed joint. When the thickness of the filler layer was $112\mu\text{m}$, it was unable to form brazed joint. In figure 3e, a lot of intermetallic compound was generated at the aluminum-steel interface. Figure 3i shows that when the transition thickness $\geq 134\mu\text{m}$. The brazed joint could be formed and intermetallic compound was not found.

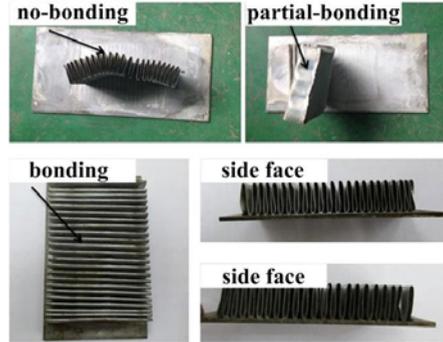


Fig.2 Schematic diagram of brazing results

Table 3 Brazing Results Of Aluminum-Steel Cladding Strip With Different Thickness Of Filler Layer And Transition Layer

Temperature(°C)	Holding time(min)	62:112(μm)	76:134(μm)	87:155(μm)
570	10	⊙	○	○
580	10	●	⊙	⊙
590	10	●	●	●
595	10	●	●	●
600	10	○	●	●

● completely bonding; ⊙ partial bonding; ○ no-bonding

When brazing temperature range was 570 °C ~580 °C, the filler cannot be fully melted. Poor liquidity of the filler led to poor brazed joint. When brazing temperature $T \geq 600$ °C and the transition layer thickness $\leq 112 \mu\text{m}$, the brazed joint cannot be formed. The brazing penetration increased with brazing temperature rising. The optimum brazing temperature range was 590 °C ~595 °C.

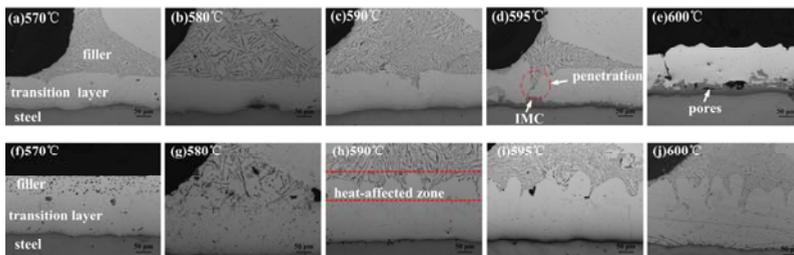


Fig.3 The Results Of Brazing With Different Thickness Of Filler Layer And Transition Layer (A-E) 62:112μm And (F-J) 76:134μm Under Different Brazing Temperature Conditions

3.2 Erosion Behavior of the Transition Layer

Table 4 Intermetallic Compound (Imc) With Different Thickness Of Filler Layer And Transition Layer

Temperature (°C)	62:112(μm)	76:134(μm)	87:155(μm)
570	□	□	□
580	□	□	□
590	■	□	□
595	■	□	□
600	■	■	□

□ without IMC, ■ IMC

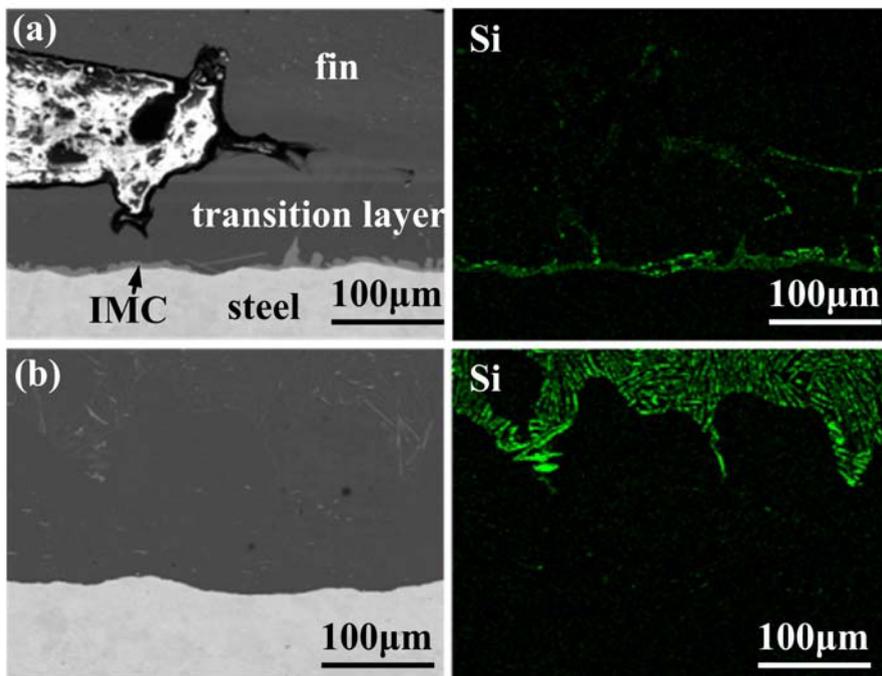


Fig.4 SEM images and Si element distribution at the brazed joint with filler layer: transition layer were (a) 62:112μm (b) 76:134μm

The Si content in the filler (7.5wt.%) was much higher than that in the transition layer (0.81wt.%). Si diffused from filler (high concentration) to transition layer (low concentration). The diffusion behavior of Si had an important influence on the erosion of the transition layer. During brazing process, the molten filler gathered in brazed joint with the aid of capillary force. The silicon's diffusion led to silicon's segregation at grain boundaries of the transition layer. The increasing of Si content led to the decreasing of the melting point of grain boundaries. When the melting point of the grain boundaries was lower than the brazing temperature, the grain boundaries melting occurred, which can produce grain boundary penetration. The phenomenon that liquid metal penetrated into the

solid metal grain boundaries, namely the grain boundary penetration[6]. The cause of penetration was that a large number of crystal defects existed in grain boundaries. The crystal lattice distortion seriously increased the energy of atoms in this region, leading to the increasing of the frequency of the atoms. Therefore, the grain boundary was conducive to the diffusion of elements[7], which was the high speed channel for the diffusion of Si.

When the transition layer thickness was 112 μm , filler penetrated transition layer along the grain boundaries of the transition layer at 600 $^{\circ}\text{C}$. Filler infiltrated to the aluminum-steel interface, transverse diffused along the aluminum-steel interface. At the same time, the increasing of filler at the aluminum-steel interface reduced melting point of the aluminum that was adjacent to steel, which led to the formation of intermetallic compounds. According to the Kirkendall effect, in figure 3e some pores appeared in the transition layer[8] due to the the diffusion rate of Al atoms was faster than Fe atoms. The silicon's diffusion from brazing filler to transition layer during brazing heating process led to form Si deficiency in filler layer, resulting in decreasing fluidity of brazing filler and more chance to form defects, such as no-bonding and pores. At the brazed joint, filler almost completely losted in figure 4a. Excessive diffusion of the filler caused pit and even perforation, which weakened the mechanical properties of the brazed joint.

When the transition layer thickness was 134 μm , the transition layer was corroded by the molten filler but not be penetrated. The molten filler diffused to the surface of the transition layer, until reached saturation. Then, the surface of the transition layer began to dissolve into the molten filler. Under the condition that the molten filler was not flowing, the molten filler first solidificated along the surface of the transition layer. Due to the transition layer corroded to a certain depht by the molten filler, the heat- affected zone formed after the filler solidification. Figure 3h-j showed that coarse $\alpha\text{-Al}$ distributed in the filler. However, interfacial compound was not found at the interface of the aluminum-steel. In figure 5, we can see that the higher the brazing temperature, the larger the heat-affected zone[9]. The increasing of Si content in transition layer can promote the formation of intermetallic compound. Therefore, in order to prevent Si diffusing into the interface of aluminum-steel effectively, the transition layer thickness was at least 134 μm .

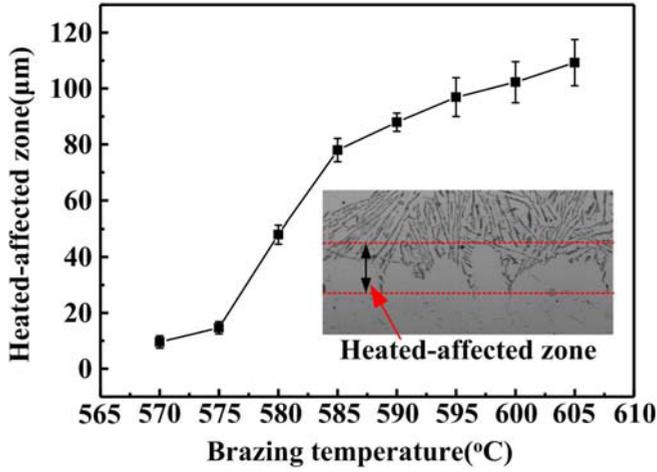


Fig.5 The Curve For Range Of Heat Affected Zone-Brazing Temperature

3.3 Intermetallic compound of aluminum-steel interface

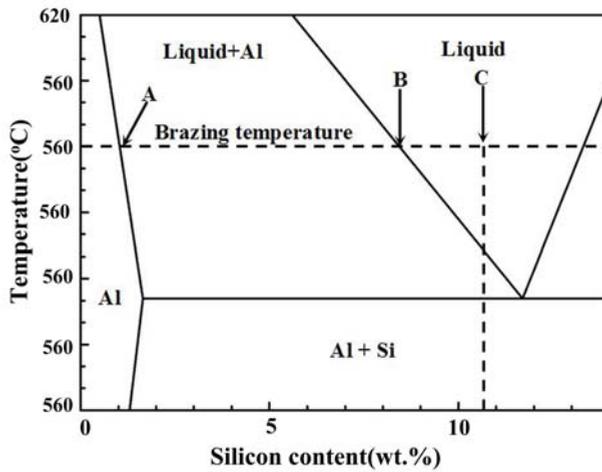


Fig.6 Al-Si phase diagram[10]

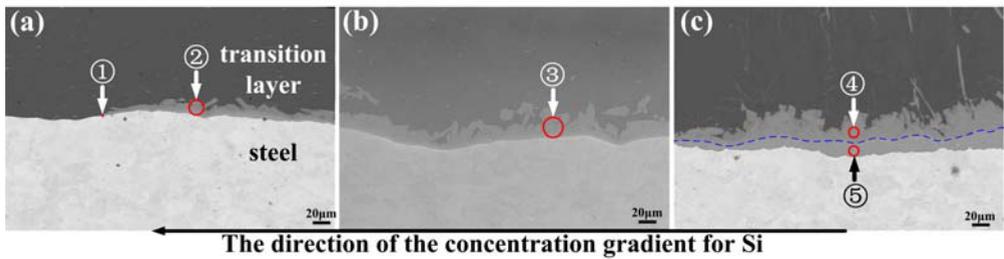


Fig.7 SEM images of the intermetallic compounds at aluminum-steel interface

Table 5 Energy Spectrum Analysis(Atom Percentage) Of Aluminum-Steel Brazing Interface

Label	B	C	D	E
Al	71.85	70.81	67.84	71.55
Si	0.91	2.63	15.31	9.85
Fe	27.24	26.56	16.85	18.6
Phase	Fe ₂ Al ₅	Fe ₂ Al ₅	Al _{4.5} FeSi	Al ₈ Fe ₂ Si

The Si in filler diffused to the interface of aluminum-steel along the grain boundary of transition layer in the form of penetration. Then Si diffused along the interface of aluminum-steel in transverse direction. The increasing of Si at the interface will increase the diffusion rate of aluminum atom and promote the formation of intermetallic compound(Figure 7a、 b).

The termination point of IMC is in ① position. IMC were embedded in the transition layer, which is a block like morphology(Figure 7b). The Si content of the ①②③ point of IMC was not much increased. At the point of ①②③, Al-Fe interdiffusion phenomenon mainly occurs. The reaction process was the following three stages[11].

In the first stage, Al-Fe interdiffused at the interface, forming a certain range of diffusion layer. The diffusion coefficient of Fe atoms in aluminum is larger than the diffusion coefficient of Al atoms in iron. There will be a large number of Fe atoms across the interface with the increasing of temperature or time. On side of the aluminum layer, Fe reacted with aluminum generating FeAl₃ (θ phase) according to Fe-Al phase diagram[12]. Al atoms continued to diffuse into the iron through FeAl₃, forming Fe-Al solid solution on the iron side. Fe atoms diffused into the transition layer through FeAl₃, forming FeAl₃ by reacting with aluminum. As a result, the intermetallic compound grew to the transition layer. The growth rate was controlled by the diffusion coefficient of Fe atoms.

In the second stage, with the increasing of the diffusion of Fe atoms, the FeAl₃ and Fe atoms reacted to form Fe₂Al₅, which is very brittle in nature. It's larger thickness is detrimental to forming properties of the material. The interface of the Fe₂Al₅ phase shows highly irregular strip shape morphology (embedded in the transition layer). The thickness of intermetallic compound increased gradually. With the diffusion of Al atoms, the reaction among Al atoms、 FeAl₃ and Fe-Al solid solution occurred: Al+FeAl₃+Fe(Al)→Fe₂Al₅. The saturation atom of Fe₂Al₅ is only 70%, and the vacancy density is high. There are a large number of vacancies in the lattice along the c axis. The Al atom can diffuse rapidly along the c axis in the Fe₂Al₅. Fe₂Al₅ grew up to form a columnar crystal region(Figure7②③).

In the third stage, the diffusion coefficient of Fe atoms in Fe₂Al₅ (η phase)is faster than that of Al atoms. As a result, Fe₂Al₅ grew to the transition layer side. The thickness of the intermetallic compound increased and thickness of the transition layer decreased. The diffusion of Fe and Al lead to the growth of Fe₂Al₅. Finally, the entire interfacial

compounds were mainly Fe_2Al_5 columnar crystals.

The intermetallic layers can be identified as τ_5 phase- $\text{Al}_8\text{Fe}_2\text{Si}$ (adjacent to steel) and τ_6 phase- $\text{Al}_{4.5}\text{FeSi}$ (adjacent to aluminum) by the EDS analysis. The initial Si content of the transition layer was 0.8 wt.%(A point in figure 6). According to the phase diagram of Al-Fe-Si, when the concentration of Si in the interface was more than A point in figure 7, which can decrease the melting point of aluminum alloy and acceleratethe IMC growth at the interface of aluminum-steel[13]. Aluminum alloy at the interface of aluminum alloy melted. The Fe_2Al_5 disappeared finally as the content of Si increasing at the interface. Figure 7c shows that in the high Si concentrations region, Al-Fe-Si compounds formed at the aluminum-steel. The results of EDS show that the two intermetallic layers were τ_5 phase($\text{Al}_8\text{Fe}_2\text{Si}$) and τ_6 phase($\text{Al}_{4.5}\text{FeSi}$), in figure 7c ④⑤.

According to the report in Ref[14]Si atom was easy to fill the Fe_2Al_5 vacancy because of the high vacancy density of Fe_2Al_5 . For this reason, interfacial compound was two layers.

Vybornov et al.[15]calculated that $\Delta_f H^0(\tau_5) = (-24.5 \pm 2)\text{kJ/mol}$ 、 $\Delta_f H^0(\tau_6) = (-34.3 \pm 2)\text{kJ/mol}$. The results showed that the formation enthalpy of phase τ_5 was the lowest, and the second is phase τ_6 . The diffusion reaction can be divided into the following two stages.

In the first stage, Si, contained in filler, diffused and penetrated into the interface of aluminum-steel. This lead to the first generation of phase $\tau_5(\text{Al}_8\text{Fe}_2\text{Si})$.

In the second stage, diffusion of Fe through phase τ_5 ($\text{Al}_8\text{Fe}_2\text{Si}$) in the transition layer and reacted with τ_5 generating phase τ_6 ($\text{Al}_{4.5}\text{FeSi}$). From the steel layer to the transition layer was τ_5 、 τ_6 , respectively.

4 Conclusions

To sum up, through the study of the brazing behavior of 3003 fin and 4343/4A60/08Al three layers aluminum-steel cladding strip used in power station air cooling system. The conclusion of this paper are listed as follows.

(1) The optimum brazing temperature range was $590^\circ\text{C} \sim 595^\circ\text{C}$ with a holding time of 10min, Meanwhile, the thickness of the transition layer must be equal to or greater than $134\mu\text{m}$.

(2) At the condition of brazing temperature less than or euqal to 580°C , the fluxility of the molten fillers was poor. Many defects were found at the brazing joint.

(3) At the condition of brazing temperature 600°C , the transition layer was seriously corroded. Moreover, when the transition layer thickness was less than or equal to $112\mu\text{m}$, the molten filler penetrated through the transition layer, resulted in the intermetallic compounds formation at the aluminum-steel interface.

(4) During brazing progress, Si in the filler diffused along the grain boundaries of the transition layer and gathered along the interface of transition layer and steel layer, leded to

the generation of intermetallic compounds. At the edge of the intermetallic compounds away from the brazing joint, the Si concentration was lower, and the intermetallic compound were mainly $\eta(\text{Fe}_2\text{Al}_3)$. In the region nearby brazing joint, with higher concentration of Si, the intermetallic compounds had a two-layer structure. One layer closing to transition layer was mainly τ_5 , while the other closing to steel layer was mainly τ_6 .

5 Acknowledgement

Authors would like to thank the support from Jiangsu Key Laboratory for Clad Materials (No:BM2014006), International Science & Technology Cooperation Program of Wuxi (No:CZE02H1504), Program on Jiangsu Science and Technology Achievements Transformation (No:BA2015079).

References

1. Li, X.Y.Zeng, X.Chen, Ch.Gao, X.J.Zhang, D.J.Zhou. Development of Aluminum-Steel Cladding Strip with Pre-Cladded Brazing Layer Used in Air Cooling System of Power Plants[J]. *Materials for Mechanical Engineering*, 2015, 39(10):21-28.
2. L.Li, N.Kotobu, F.X.Yin. Progress in cold roll bonding of metals[J]. *Science and Technology of Advanced Materials*, 2008, 9(2):1-11.
3. H.D.Manesh, H.S.Sha. Effective parameters on bonding strength of roll bonded Al/St/Al multilayer strips[J]. *Journal of Alloys and Compounds*, 2009, 476(1/2): 292-299.
4. D.L.Chen. The Study on Aluminum-steel Plate-fin Heat Exchanger Brazing Technology[D]. Lanzhou University of Technology, 2014.
5. F.N.Afshar, J.H.W.de Wit, H.Terryn, J.M.C.Mol. The effect of brazing process on microstructure evolution and erosion performance of a modified AA4XXX/AA3XXX brazing sheet[J]. *Erosion Science*[M], 2012(58):242-250.
6. Okamoto,T.Takemoto, K.Uchikawa, Brazability and Erosion by Molten Filler Alloy of Aluminum Base Plates with Intermetallic Compounds[D]. Osaka University, 1983.
7. D.J.Schmatz. Grain Boundary Penetration During Brazing of Aluminum[J], *Welding Journal*, 1983, 62(10): 267-271.
8. D.J.Zhou, Zh.Chen, X.J.Zhang, J.G.Tang. Effect of Si Addition on Intermetallic Compound Layer at Aluminum/Steel Interface Bond by Rolling[J]. *Heat Treatment of Metals*, 2014, 39(1):456-468.
9. A.W.Ralph. Liquid film migration during aluminum brazing[J]. SAE paper 971848,1997.
10. J.L.Murray, A.J.McAlister. The Al-Si(aluminum-silicon)system[J]. *Bulletin of Alloy Phase Diagram*, 1984, 5(1):74-84.

11. J.Vikas, V.C.Srivastava, D.Arpan, R.N.Ghosh. Reactive diffusion in the roll bonded iron-aluminum system[J]. *La Metallurgia Italiana*, 2006(7/8):55-59.
12. H.Okamoto. Phase Diagrams of Binary Iron Alloys[M]. Materials Park, OH: ASM International, 1993.
13. M.Yousaf, J.Iqbal, M.Ajmal. Variables affecting growth and morphology of the intermetallic layer(Fe_2Al_5)[J]. *Materials Characterization*, 2011:517-525.
14. T.Heumann, M.Dittrich. Structure character of the Fe_2Al_5 intermetallic compound in hot dip aluminizing process[J]. *Z Metallk*, 1959, 50:617-623
15. M.Vybornov, P.Rogl, F.Sommer F. On the thermodynamic stability and solution behavior of the phases- $\text{Al}_{7.4}\text{Fe}_2\text{Si}$ and τ_6 - $\text{Al}_9\text{Fe}_2\text{Si}_2$ [J]. *Journal of Alloys and Compounds*, 1997, 247(1/2): 154-157.