Journal of Nuclear Materials 395 (2009) 145–149

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

High strength bonding of titanium to stainless steel using an Ag interlayer

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ARTICLE INFO

Article history:
Received 3 March 2009
Accepted 7 October 2009

ABSTRACT

Strong bonding between titanium (Ti) and stainless steel (STS) was achieved by employing a commercially available Ag–28Cu (wt.%) filler, and more importantly with a help of an Ag interlayer. A mass transport of the Ti elements from the substrate into the molten filler was completely prevented through the use of the Ag interlayer, so the resultant brazed joint was free from any brittle Ti-based intermetallic compounds. Notably, this Ti–STS dissimilar joint displayed a remarkable improvement in its bonding strength, demonstrating the potential application of an Ag interlayer for joining Ti and its alloys to various structural steels.

1. Introduction

Titanium (Ti) is the material of choice for demanding corrosive environments in nuclear industry due to its high specific strength and excellent corrosion resistance [1,2]. With the expanding use of Ti, there is a great demand for joining of Ti to more commonly used stainless steels (STS) [3,4]. This requirement is not always caused by the cost, but also by the desire for yielding an enhanced corrosion resistance for a number of nuclear structural components. For example, Ti is the natural choice as a dissolver material in a nuclear fuel reprocessing plant, while various processing equipment and piping in the plant are made of STS [1].

Despite such a strong need for Ti–STS transition joints, it is generally known to be very difficult to produce a reliable and strong joint due to a lack of metallurgical compatibility, and the formation of intermetallic compounds (IMCs) between these two materials [5,6]. Although there have been many studies on a dissimilar joining of Ti to STS by various processes including fusion welding, diffusion bonding and brazing, most of the joints have revealed poor bonding characteristics due to the formation of brittle Ti-based IMCs such as Ti_xFe_y, Ti_xCu_y, Ti_xNi_y and Ti_xCr_y [7–9]. Those IMCs were unavoidably produced by the reaction of the Ti elements coming from the substrate with either the counterpart STS substrate or the fillers used [5–9]. Additional efforts have also been made to avoid such a fundamental brittleness of a Ti–STS dissimilar joint by using a diffusion barrier such as Ni, Cu and Cr, but they were unsuccessful in suppressing the formation of the brittle IMCs, owing to an undesired reaction of the diffusion barrier with either the substrates or the fillers used [10–12].

In this study, we report on a strong and robust metallurgical bonding between Ti and STS by simple brazing with a BAg-8 (Ag–28Cu (wt.%)) filler, which is most widely utilized for brazing of Ti and its alloys. Of more technological importance, this was achieved by employing a suitable diffusion barrier, i.e., Ag interlayer, on the Ti substrate. The Ag interlayer was chosen as a diffusion barrier by considering its interfacial reactions with both the Ti substrate and the BAg-8 filler. First, the Ag interlayer was expected to be an effective barrier to a mass transport of the Ti substrate into the molten filler without forming any brittle reaction compounds at the Ti interface. It was also expected that the Ag interlayer would not only be chemically compatible, but also be irrelevant to the formation of brittle IMCs with the BAg-8 filler [13].

2. Experimental procedure

Square rods of Ti (Gr.2) and STS (UNS31254) with a size of 10 mm × 10 mm × 15 mm were used as the substrates in this study. UNS31254 was a super stainless steel containing 20Cr–18Ni–6Mo (wt.%). A commercially available Ag–28Cu (wt.%) eutectic alloy foil (BAg-8) with a thickness of about 70 μm was used as the filler material. The total area of the filler was approximately the same as that of the substrate.

The Ag interlayer was coated onto the Ti substrate by a sputtering technique. The sputtering pressure was maintained at 0.8 Pa in a pure argon atmosphere. The produced Ag layer was uniform with a thickness of approximately 40 μm. The Ag layer with a thickness of 20 μm was also prepared to verify the effect of the interlayer thickness on the brazability.

The infrared brazing technique was used for the Ti–STS dissimilar joining. Prior to brazing, the infrared chamber was evacuated down to 7×10^{-3} Pa. Infrared brazing was then performed at 790–830 °C for 5–10 min. A constant (4 l/min) flow of argon was

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doi:10.1016/j.jnucmat.2009.10.045

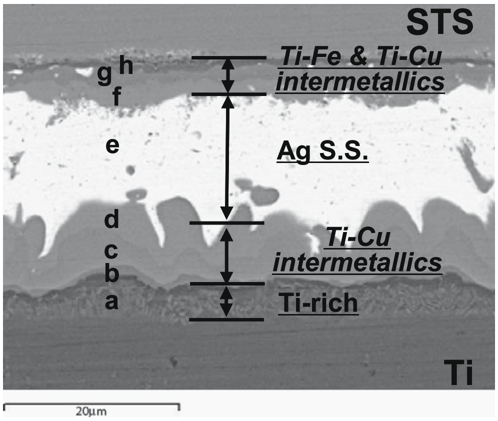

Fig. 1. SEM BEI of the joint brazed at 810 °C for 10 min without an Ag interlayer.

supplied during brazing. The heating and cooling rates were maintained at 100 °C/min throughout the experiments.

The cross-sections of the brazed joints were examined using a scanning electron microscope (SEM). Quantitative chemical analyses of the joints were also performed using an electron probe microanalyzer (EPMA) with a spot size of 1 μm. In order to evaluate the bonding strength of the Ti–STS dissimilar joints, room-temperature tensile tests were conducted at a strain rate of $8.3 \times 10^{-4}\ s^{-1}$ using samples with a gauge length of 10 mm. After the tensile tests, the cross-sectional areas adjacent to the fractured surfaces were investigated using an SEM.

3. Results and discussion

Fig. 1 shows the SEM back-scattering electron image (BEI) for the Ti–STS dissimilar joint brazed at 810 °C for 10 min with a BAg-8 filler alone, and the relative chemical analysis results by the EPMA are presented in Table 1. It was generally observed that the dissolution of the Ti substrate was much more prominent as compared to that of the STS substrate, and the resultant joint was mainly involved in an intensive reaction of the Ti elements dissolved from the substrate with the ones coming from both the molten filler and the counterpart STS substrate. As shown in Fig. 1, those Ti elements preferentially reacted with the Cu in the molten filler to form thick Ti_2Cu, TiCu and Ti_3Cu_4 IMC layers (b–d in Fig. 1) near the Ti substrate. In part, they also diffused into the molten filler and produced a relatively thin TiFe IMC layer near the STS interface by reacting with the STS substrate (h in Fig. 1). The Ag elements in the molten filler were mostly segregated at the center of a joint (e in Fig. 1) and had a negligible effect on the interfacial reactions with both the Ti and STS substrates.

Like those in previous studies [8,9,11,14], the Ti–STS brazed joint was dominated by brittle Ti-based IMCs due to a significant dissolution of the Ti substrate. It was also considered that these Ti-based IMCs could not be avoided by only using a Ag-based filler (BAg-8) alone in our present experimental regime. By recognizing this, an additional Ag interlayer was introduced as a diffusion barrier to the dissolution of the Ti substrate. If the dissolution of the Ti substrate could be completely prohibited by the Ag interlayer, then only the Ag and Cu elements would be present in the molten filler. Consequently, it was reasonable that the resultant joint would be evolved from an Ag–Cu eutectic reaction of the molten Ag–Cu liquid, i.e. $[L \rightarrow (Ag) + (Cu)]$ at the cooling stage, thus not involving any reactions to form brittle IMCs [13]. Furthermore, it was expected that no IMCs would be formed at the STS interface, as noted from the Ag–(Fe,Ni,Cr) and Cu–(Fe,Ni,Cr) binary alloy phase diagrams [13]. At the Ti interface, a TiAg phase might be formed as a result of an interfacial reaction between the Ti substrate and the Ag interlayer [13], but it has not been considered as brittle as most other Ti-based IMCs, nor has it been found to deteriorate the bonding strength of a joint, as demonstrated by previous studies [14–16].

Such predictions were in good agreement with the experimental observation, as shown in Fig. 2. In contrast to the sample brazed with the BAg-8 filler alone (Fig. 1), the joint consisted of only pure Ag and Ag-rich solid solution regions, which was completely free from any brittle Ti–Fe and Ti–Cu IMCs. As expected, the only observed interfacial reaction compound was the TiAg phase, which was uniformly produced along the boundary of the Ti substrate with a thickness of about 15 μm (a in Fig. 2a). The absence of the Ti-based IMCs suggested that a mass transport of the Ti elements from the Ti substrate into the molten filler was completely prevented by the presence of the Ag interlayer. At the STS interface, a thin diffusion zone of the main elements (Ag, Fe, Ni, and Cr) was produced without any reaction compounds (Fig. 2b). Smooth and continuous variations of the elemental concentration profiles in Fig. 2b also indicated a strong diffusion bonding between the STS substrate and the Ag-rich solid solution.

It was unpredictable, however, that there was no Ag–Cu eutectic structure produced by the $[L \rightarrow (Ag) + (Cu)]$ reaction in the whole joint. This could be explained by an isothermal solidification of the Ag–Cu molten filler into the Ag-rich solid solution, as shown in Fig. 3. At the early stage of brazing, a compositional change of the molten filler by a dissolution of the Ag interlayer (arrow A in Fig. 3) occurred in the vicinity of the Ag interlayer, resultantly initiating the isothermal solidification of the molten filler. As the isothermal holding continued, this isothermal solidification proceeded by a diffusion of the Cu elements into the Ag interlayer (explained by points c–f in Table 2), as accompanied by a compositional change (arrow B in Fig. 3) from a molten Ag–Cu liquid (x in Fig. 3) to a solidifying solid (y in Fig. 3) at a solid/liquid interface. When the isothermal holding time was long enough for all the molten filler to be solidified into the Ag-rich solid solution, the isothermal solidification virtually led to a joint structure of Ti(substrate)/TiAg/Ag/Ag S.S./STS(substrate) as shown in Fig 2a.

As described, the isothermal solidification of the Ag–Cu molten filler into the Ag-rich solid solution was rate-controlled by the diffusion of the Cu elements into the Ag interlayer. Experimentally, an insufficient isothermal holding time (e.g., 5 min at 810 °C) induced the formation of the Cu-rich phases near the STS interface due to

Table 1
EPMA results of the marked areas in Fig. 1 for the brazed joint without an Ag interlayer.

	Ti (at.%)	Ag (at.%)	Cu (at.%)	Fe (at.%)	Cr (at.%)	Ni (at.%)	Mo (at.%)	Phase
a	93	1	6	–	–	–	–	Ti-rich
b	66	2	32	–	–	–	–	Ti_2Cu
c	51	3	46	–	–	–	–	TiCu
d	42	2	56	–	–	–	–	Ti_3Cu_4
e	–	92	8	–	–	–	–	Ag S.S.
f	30	7	54	3	–	6	–	$TiCu_2$
g	41	–	43	10	2	4	–	TiCu
h	36	–	8	35	13	6	2	TiFe

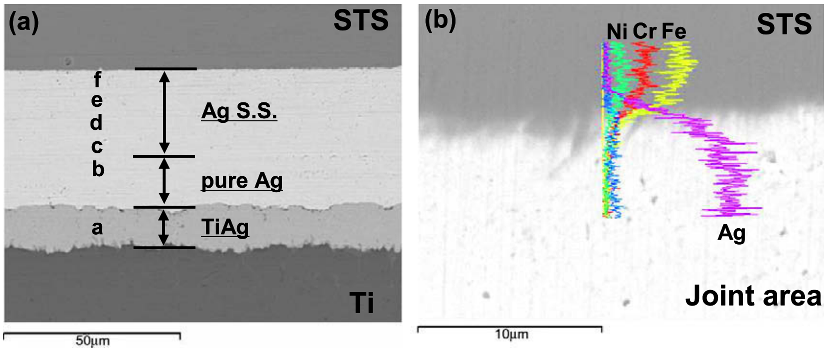

Fig. 2. SEM BEIs of the joint brazed at 810 °C for 10 min with an Ag interlayer; (a) the whole joint and (b) a close view near the STS interface.

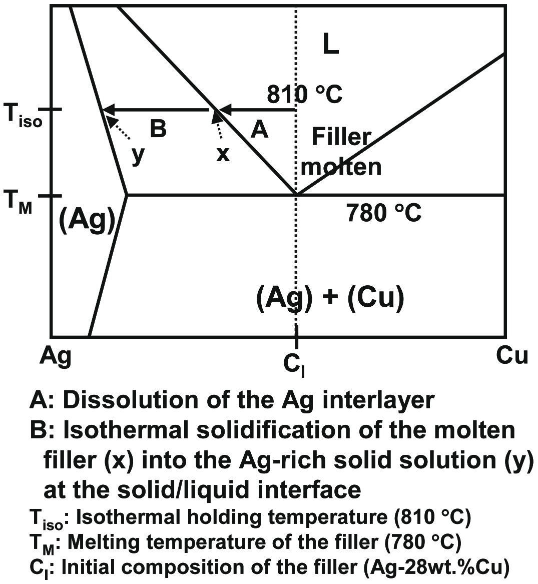

Fig. 3. Compositional changes of the constituent phases during Ti–STS dissimilar brazing using both an Ag–28Cu (wt.%) filler and an Ag interlayer.

Table 2
EPMA results of the marked areas in Fig. 2a for the brazed joint with an Ag interlayer.

	Ti (at.%)	Ag (at.%)	Cu (at.%)	Phase
a	50	50	–	TiAg
b	–	100	–	Ag
c	–	99	1	Ag S.S.
d	–	97	3	Ag S.S.
e	–	95	5	Ag S.S.
f	–	94	6	Ag S.S.

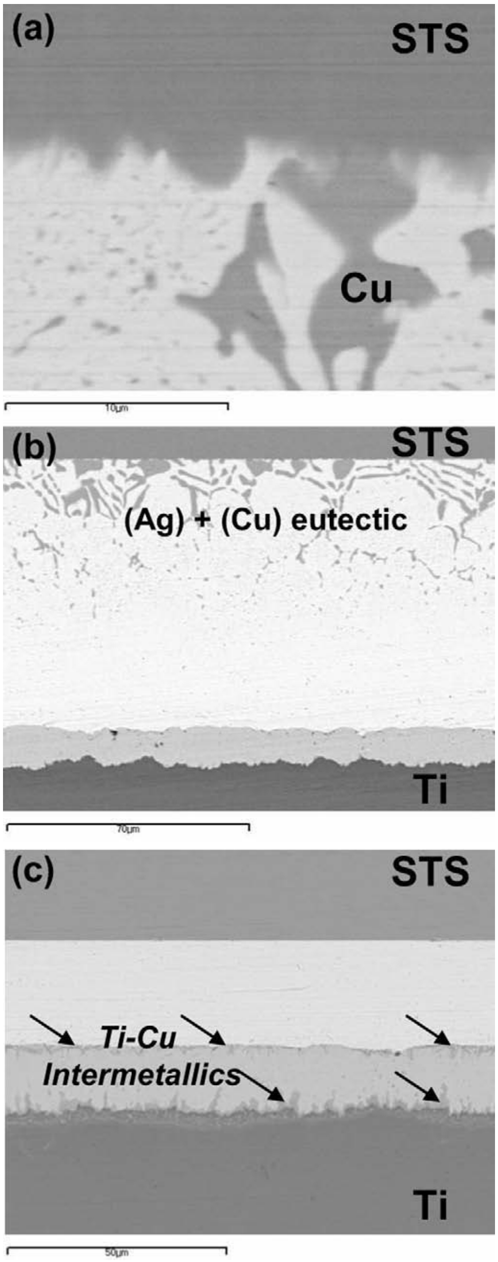

Fig. 4. SEM BEIs of the joints brazed at (a) 810 °C for 5 min (a close view), (b) 790 °C for 10 min and (c) 830 °C for 10 min with an Ag interlayer.

the limited diffusion of the Cu elements (Fig. 4a). Furthermore, the distinctive (Ag) + (Cu) eutectic microstructure was observed when the brazing temperature decreased to 790 °C, as shown in Fig. 4b. At such low temperatures, the isothermal solidification rate was so low that a considerable amount of the molten Ag–Cu liquid still remained after the isothermal holding period. As a result, the (Ag) + (Cu) microstructure was produced by the eutectic solidification of the remaining molten filler during the subsequent cooling. As the brazing temperature increased to 830 °C contrarily, the diffusion rate of the Cu elements in the Ag interlayer was too high to prevent the Cu elements from encountering the Ti substrate, thereby resulting in the formation of Ti–Cu IMCs at the Ti interface (indicated by arrows in Fig. 4c). The brazing temperature should be lower than 820 °C to prohibit such IMC formation when using the Ag interlayer with a thickness of 40 µm.

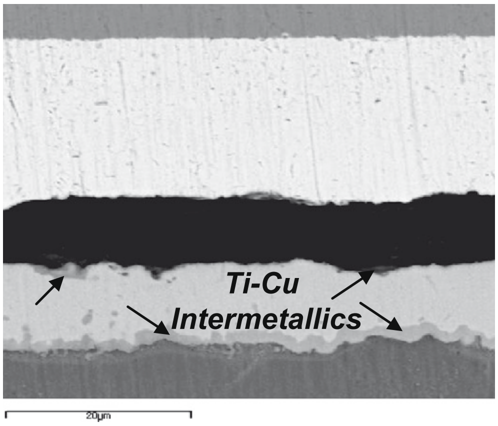

Fig. 5. SEM BEI of the joint brazed at 810 °C for 10 min using an Ag interlayer with a thickness of 20 μm.

Fig. 5 shows the SEM BEI for the Ti–STS joint brazed at 810 °C for 10 min using an Ag interlayer with a thickness of 20 μm. As shown in the figure, the permanent crack was observed between the Ag interlayer and the continuously formed TiAg phase. In this case, the thickness of the Ag interlayer (20 μm) was insufficient to obstruct the diffusion of the Cu elements into the Ti substrate, so the detrimental Ti–Cu IMCs (arrows in Fig. 5) were produced by the reaction with the Ti elements from the substrate. These interfacial Ti–Cu IMCs acted as fracturing sites during the cooling stage due to their thermal mismatch with the adjacent phases. In our study, the Ag interlayer should be thicker than 30 μm to manifest the effectiveness of the Ag interlayer as the diffusion barrier, indicating that the diffusion rate of the Cu elements in the Ag interlayer was about 3 μm/min at this brazing temperature.

Fig. 6 shows the stress–strain curves obtained by the room-temperature tensile tests for the samples brazed at 810 °C for 10 min with and without an Ag interlayer, and the corresponding fracture images are also shown in the insets. To the best of our knowledge, the bonding strengths of the Ti–STS dissimilar joints are known to be too low (usually less than 100 MPa) for these joints to be technically usable due to the presence of brittle Ti-based IMCs [8,9,12]. In some cases, higher strengths (up to 250 MPa) have been obtained by optimizing a joint structure [11,17]. It is worthwhile noting, however, that all the previous studies failed to suppress the formation of brittle IMCs in a joint, and the resultant bonding strengths were not reproducible, owing to a lack of structural consistency for the brittle IMCs from sample to sample. Likewise, the samples brazed with the BAg-8 filler alone in this study were fractured at an early stage before yielding at a relatively low fracture strength of about 60 MPa, revealing a brittleness of a joint (Fig. 6). The fractured samples also revealed that a crack propagated along a brittle TiFe IMC layer, as shown in the lower inset of Fig. 6.

On the contrary, the bonding strength of the Ti–STS dissimilar joint was remarkably improved, when the formation of brittle Ti-based IMCs was completely suppressed by the application of an Ag interlayer. The strengths at the fractures were measured to be about 410 MPa with a high reproducibility. It was also obvious that yielding of the substrate occurred before a fracture as shown in Fig. 6. When compared to the samples without an Ag interlayer, the fracture occurred along the pure Ag or the Ag-rich solid solution region, as shown in the upper inset of Fig. 6. Notably, the continuously produced TiAg layer remained quite stable after the tensile tests and did not deteriorate the bonding strength of the Ti–STS dissimilar joint.

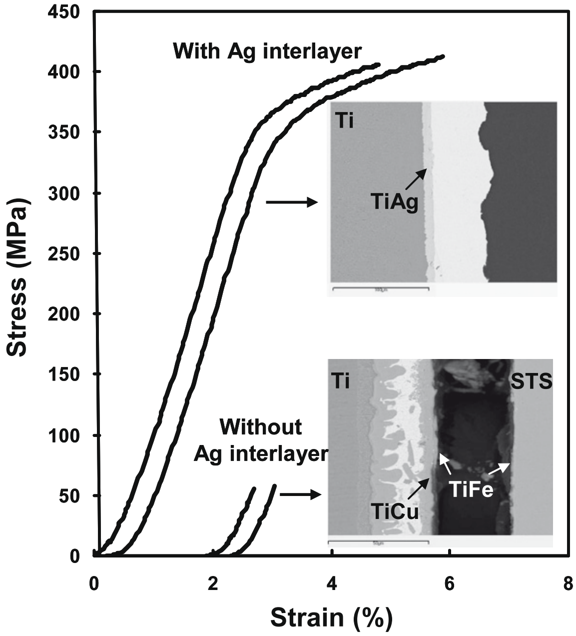

Fig. 6. Tensile stress–strain curves for the samples brazed at 810 °C for 10 min. Insets show the cross-sectional areas adjacent to the fractured surfaces.

Table 3
Tensile bonding strengths of the Ti–STS joints brazed using an Ag interlayer.

Brazing temperature (°C)	790	810	830
Bonding strength (MPa)	405 ± 10	410 ± 10	150 ± 45

Table 3 shows the effect of the brazing temperature on the bonding strength of the Ti–STS joints brazed using an Ag interlayer. It was noted that the bonding strength of the joint brazed at 790 °C was almost same as the joint brazed at 810 °C. This result revealed that the diffusion reaction at both the Ti and STS interfaces was sufficient to obtain the strong bonding even at this low brazing temperature. However, the high brazing temperature (830 °C) impaired the bonding strength of the joint owing to the detrimental Ti–Cu IMC formation near the Ti interface as shown in Fig. 4c. It is not shown here, but the fracture occurred along the interface between the Ag interlayer and the TiAg phase in the samples brazed at 830 °C.

4. Conclusion

In this study, a strong joining of Ti to STS was achieved by brazing with a BAg-8 filler, particularly with the help of an Ag interlayer on the Ti substrate. The Ag interlayer effectively prevented a mass transport of the Ti substrate into the molten filler during brazing, thereby producing a highly reliable joint structure of Ti(substrate)/TiAg/Ag/Ag S.S./STS(substrate) without any detrimental IMCs. Notably, the bonding strength of the resultant Ti–STS dissimilar joint was remarkably improved up to 410 MPa, exceeding the yield strength of the Ti substrate.

Acknowledgement

This research was supported by a grant from the "Development of low temperature solid state joining technology of dissimilar metals for nuclear heat exchanger tube components" project funded by the Ministry of Knowledge Economy (MKE) of Korea.

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