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Recrystallization and grain growth at the interface of a bimetallic colaminated strip composed of two different Fe-Ni alloys

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Abstract. Roll bonding is a solid-state welding process widely used to manufacture layered metal composites. Particular properties may thus be obtained using the physical features of each material of the composite. Bimetal plates consisting of two different Fe-Ni alloys were made by roll bonding followed by heat treatment for 90 minutes at various annealing temperatures. The effects of post-rolling heat treatments on the bonding strength of a bimetal strip were investigated in relation to the interface microstructure evolution. Both recrystallization and grain growth took place at the interface during annealing. In particular, nucleation of new grains as well as growing grains crossing the interface may have contributed to the improvement of the bonding strength. Moreover, diffusion through the interface was found to drastically enhance the bonding strength from 850°C up to 1050°C. However, excessive grain growth associated to porosity occurrence probably caused the saturation of the bonding strength beyond 1050°C.

1. Introduction

Recent decades have been marked by the increasing production of clad metal materials. Such multi-materials aim to either save precious metal like electromechanical relays with gold-plated contacts or to achieve functional properties by combining the physical features of each material, like aluminum cookware clad with stainless steel exhibiting high heat conductivity and food compatibility. One of the most widely used techniques to make the materials strongly connected is the cladding process, commonly obtained by co-rolling of two (or more) solid-state metals at high temperature (like hot rolling) or low temperature (preventing any surface oxidation). This paper deals with the latter technique, which can also be called “Roll bonding”. One of the industrial applications is bimetal composed of two components with very different thermal expansion behaviour clad together, that bends during overheating in case of current excess in electrical devices and thus interrupts the circuit [1].

It is generally supposed that bonding mechanisms rely on the film theory, first established by Vaidyanath and Milner [2], stating that the breaking of the surface layer is first required to bring “fresh” (which said out of any surface oxidation) metals into contact at the interface. This model called “fracture – extrusion – welding” was then refined by Bay [3] who proposed that surface expansion upon rolling breaks a contaminant film and a work-hardened surface layer produced by surface preparation. Due to roll pressure, fresh metal from each opposite material can extrude through



the cracks and bond at the interface. In recent work, many parameters affecting welding quality have been reported [4]: surface preparation, thickness reduction rate, rolling temperature, deformation speed and normal pressure. Furthermore, post-heat treatments carried out after rolling usually provide an improvement in bonding strength, attributed to diffusion and short-range atomic movements [4]. However, heterogeneities induced by the rolling process may also affect bonding reinforcement during annealing. In this study, annealing treatments were performed on bimetallic strips of two iron-nickel alloys, to improve understanding of thermally activated consolidation mechanisms.

2. Experimental Method

Two different iron-nickel alloys Fe-47.5Ni and Fe-50Ni-8.6Cr (wt %), with an initial grain size of approximately 17 μm and 7 μm respectively, were used. These initial recrystallized laminations with a dimension of 75 x 10 x 1 mm³ were first degreased with acetone. The surfaces of the sheets were then scratch-brushed in order both to remove contaminants and oxides and to form a work-hardened surface layer. After riveting the heads of the sheets, they were heated at 350°C for 15 minutes in air and rapidly roll bonded right out of the oven. Rolling was performed at a thickness reduction of 50% using a laboratory rolling mill with a capacity of 2.2 kW. The rolls diameter was 67 mm and the rolling speed was 48 mm/s. After rolling, the composite strips were annealed in air for 90 minutes at different temperatures ranging from 350°C to 1200°C. Bonding strengths were measured by the peeling test [5]. The peeling test was carried out using a Zwick/ Roell Z010 tensile testing machine with a crosshead speed of 100 mm/min and a 10 kN load cell. Zeiss Sigma HD scanning electron microscope (SEM) equipped with energy dispersive spectrometer (EDS) was used to analyze the interfacial bonding state under different annealing conditions with a step size of 0.5 μm . The microstructural interfacial evolution was monitored by the electron backscatter diffraction (EBSD) technique with a step size ranging from 50 nm to 0.5 μm . Microstructural investigations were performed in the transversal section which contained the transversal (TD) and normal (ND) directions. All EBSD orientation maps are ND-IPF.

3. Results and discussion

Figure 1 presents the experimental bonding strength as a function of annealing temperature. At low temperatures, the annealing treatment shows little impact on the bonding strength, which remains steady until 550°C. From 550°C, a significant growth of adhesion is achieved with the increase of temperature. Thus, the bonding strength goes up by a factor of 30 before reaching a plateau at 1050 °C. The influence of microstructural evolution at the interface on bonding strength was then investigated. The effects of annealing temperature on the recrystallized area fraction f_{rex} and the recrystallized average grain size were also plotted in Figure 1. It appears that recrystallization occurs from 550°C and is complete at 650°C and 750°C for Fe-47.5Ni and Fe-50Ni-8.6Cr respectively (Figure 1a). Concerning the grain size, grain growth occurs as soon as recrystallization begins, at 550°C. A classical grain growth law is followed [6].

Figure 2 displays EBSD orientation maps carried out at the bonding interface of samples annealed at different temperatures. Before annealing, the microstructure of both alloys exhibits refined and elongated grains, as shown in Figure 2a. The coarse particles and non-indexed regions observed along the interface were found to be fragments of the work-hardened layer produced by scratch-brushing, broken and scattered during roll bonding. However, the welded interfacial areas were sufficient in number to ensure minimal bonding strength and allow the peeling test. For temperatures lower than 450°C, the grains show similar morphology to those of the roll-bonded sample. From 550°C, the area fraction of recrystallized grains in both components starts to rise rapidly with increasing temperature (see in Figure 1a). As recrystallization proceeds, substantial twinning occurs. The Fe-47.5Ni and Fe-50Ni-8.6Cr alloys achieve complete recrystallization at 650°C (Figure 2b) and 750°C (Figure 2c) respectively. The resulting microstructure comprises equiaxed grains, as it can be seen in Figure 2d. It is worth noting that the beginning of the increase in bonding strength coincides with the onset of grain

growth. From 650°C, both Fe-47.5Ni and Fe-50Ni-8.6Cr alloys undergo grain growth, reaching an average grain size of 28.5 and 14 μm respectively (see in Figure 1b, Figure 2d).

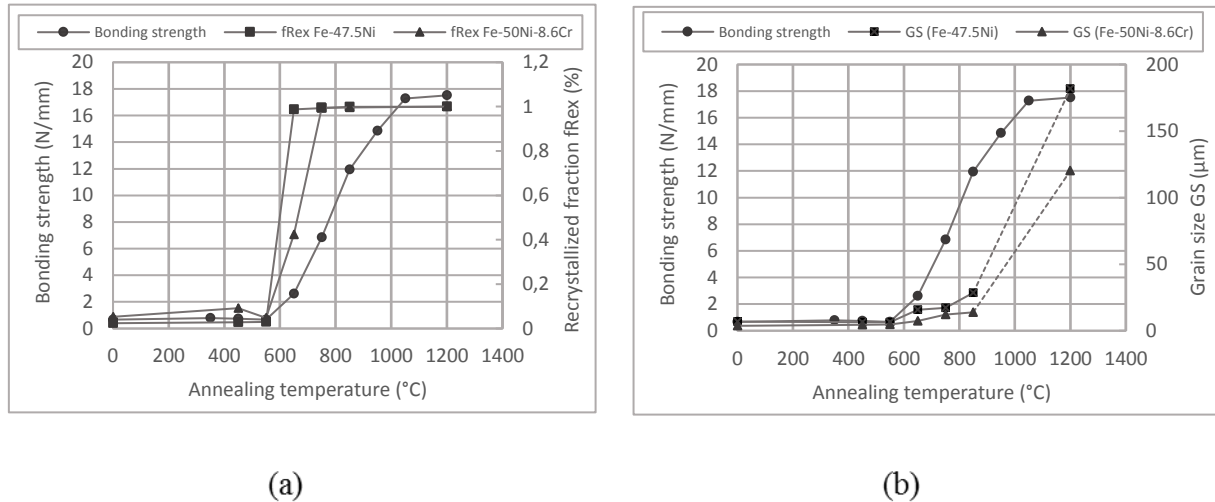


Figure 1. The effect of the annealing temperature on the bonding strength and: (a) on the recrystallized area fraction; (b) on the average recrystallized grain size of each alloy. Both the area fraction and the grain size were measured in the base alloys at some distance of the interface. Due to the small number of grains analysed, the grain size in the samples annealed at 1200°C was estimated.

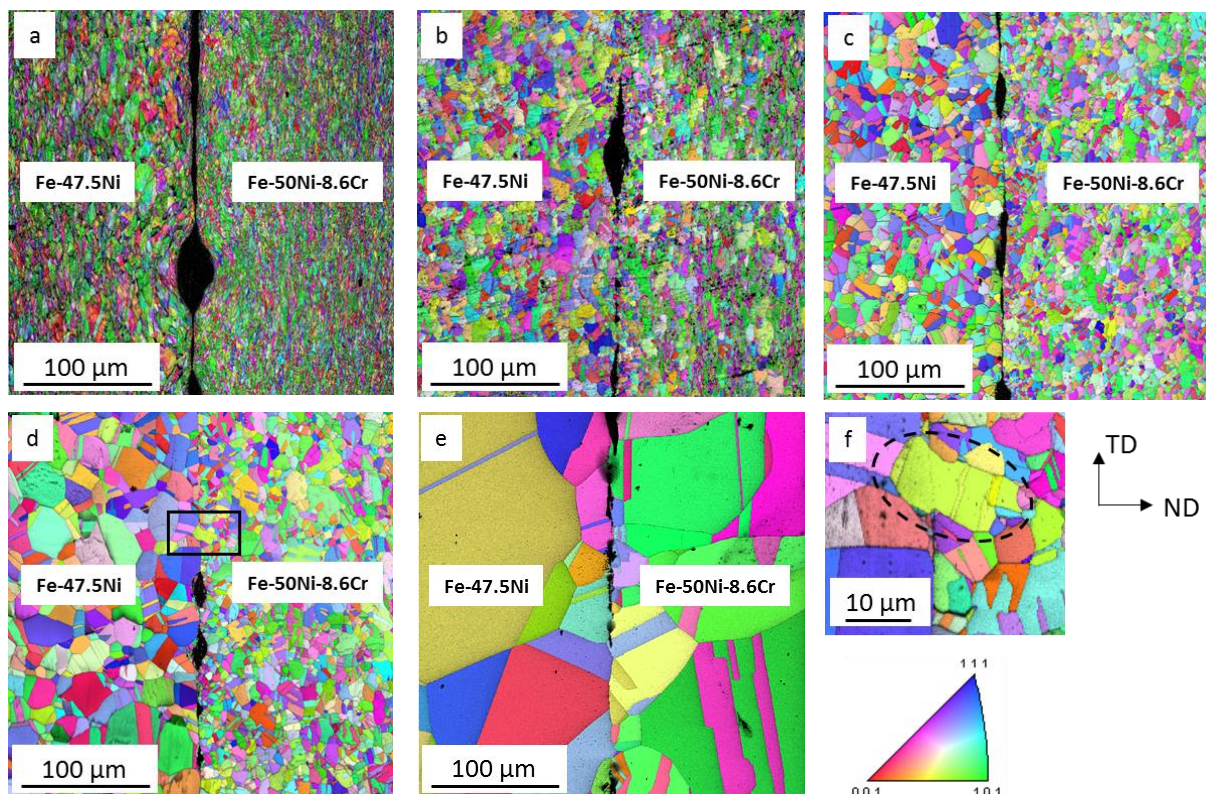


Figure 2. EBSD orientation maps showing the microstructure of the samples after 90 min of annealing at different temperatures: (a) deformed state; (b) 650°C; (c) 750°C; (d) 850°C; (e) 1200°C; (f) the box at the interface of the sample annealed at 850°C with higher magnification.

Figure 2f shows with higher magnification a part of the orientation map made at the interface of the sample annealed at 850°C (see in Figure 2d). The dotted circle points a grain which has grown and crossed the interface during annealing. Inside this grain, a ghost line [7] can be seen, marking the location of the interface before annealing.

Figure 3 displays EBSD orientation maps focused on the particles located along the interface of as-rolled and annealed samples. Similar particles arising from wire-brushing surface treatments were observed by Mishin et al. [8] at the interfaces of nickel processed by ARB. The authors found out that these work-hardened fragments affected the progress of recrystallization by providing highly refined deformation zones acting as preferential nucleation sites. This is in agreement with Figure 3a and Figure 3b, where it can be seen that these particles affect the rolled microstructure around them, creating deformation zones which contain finer grains than in the core.

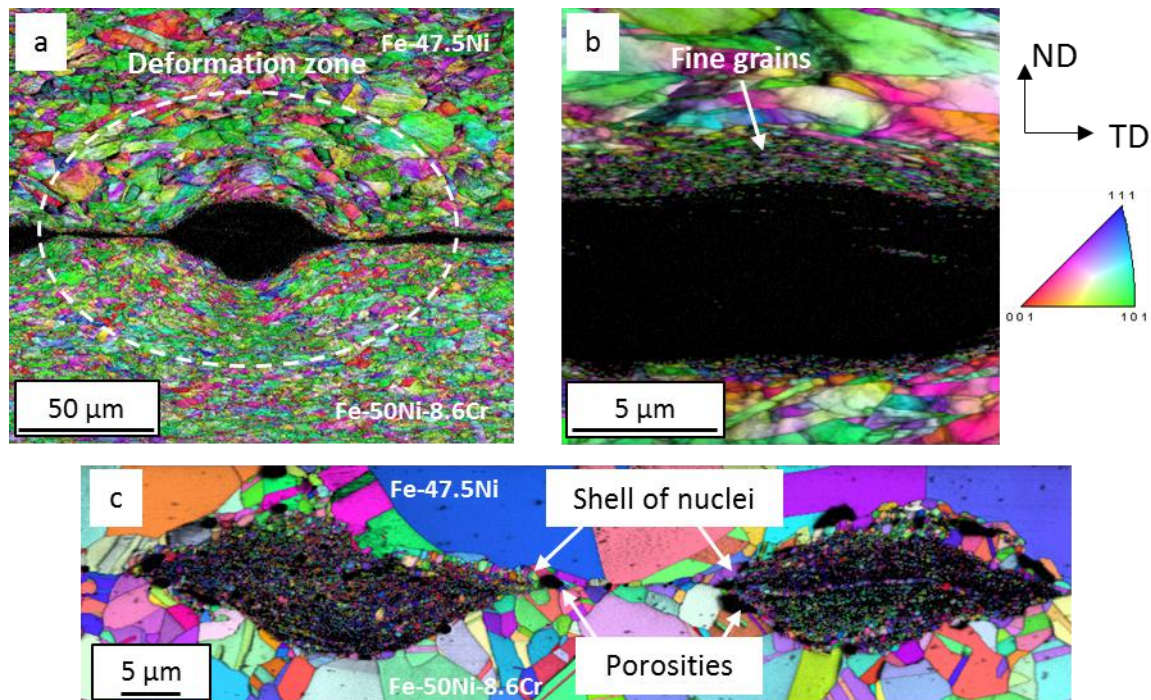


Figure 3. EBSD orientation maps showing particles at the interface of composite sheets: (a) before annealing; (b) before annealing with higher magnification; (c) after annealing at 850°C.

In Figure 3c, a “shell” made of nucleated recrystallized grains with various crystallographic orientations can be noticed around the fragments. It should be noted that regions containing fragments were recrystallized after annealing at 850°C but are composed of smaller grains than in the core of the sample. Recrystallization around fragments and interface contributed to the “interface closure” observed at larger scale in the maps of Figure 2 as annealing temperature increased (compare Figure 2a and 2d). Some porosities can be noticed around the particles after annealing at 850°C (see in Figure 3c). These voids were indeed observed along the interface of every annealed sample and seemed to become more frequent with increasing temperature.

Figure 4 shows EDS concentration profiles of major elements across the bonding interface for samples annealed at 450°C and 1200°C. As seen on the profiles, an interdiffusion layer is established at the interface and becomes larger with increasing temperature. The diffusion coefficient of Ni into Fe-52 %Ni was calculated with equation (1):

$$D(T) = D_0 \exp\left(-\frac{Q}{RT}\right) \quad (1)$$

where D_0 is the diffusion factor, Q is the activation energy for diffusion, R is the gas constant ($R = 8.314 \text{ J.mol}^{-1}.\text{K}^{-1}$) and T is the annealing temperature.

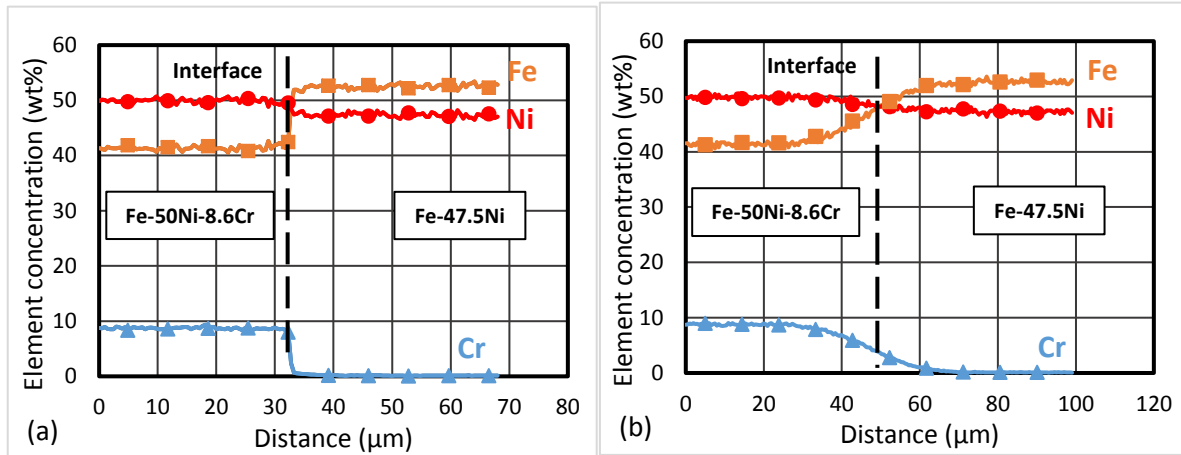


Figure 4. EDS line scanning curves of interfacial zone after annealing at: (a) 450°C; (b) 1200°C.

The theoretical variations of the interdiffusion layer thickness $L_D = \sqrt{D(T)t}$ (using equation (1)) and the bonding strength are plotted versus annealing temperature in Figure 5. The diffusion thickness is very weak at 450°C and is expected to remarkably expand at 1200°C (around 12 μm). This is in accordance with the experimental diffusion distance of Ni found from EDS measurements: 16 μm at 1200°C (Figure 4b).

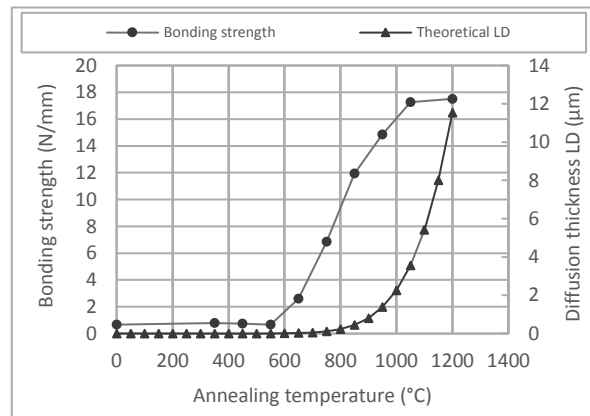


Figure 5. The effect of the annealing temperature on the bonding strength and the diffusion layer thickness of nickel at the interface of the composite sheet.

The annealing treatment performed after roll bonding involved several phenomena highlighted in this study, contributing to the bonding strength improvement of the composite with increasing temperature. Both recrystallization and grain growth driven by grain boundary migration took place during annealing. As seen previously in Figure 1b, grain growth may have promoted the increase in bonding strength from 650°C. One can assume that nucleation of recrystallized grains around coarse work-hardened fragments and at the interfacial regions contributed to enhance the bonding strength. Besides, at high annealing temperatures, some growing grains were able to cross the interface undoubtedly reinforcing bonding (see in Figure 2f).

Moreover, the observed atomic diffusion between the two sheets at high temperatures may have improved the metallurgical bonding at the interface, through a solid-solution strengthening effect. A similar process leading to an increase of mechanical bonding strength was described by Hwang et al.

[9] after annealing of an aluminum/stainless steel bimetal plate. It is suggested that volume diffusion during annealing not only resulted in strengthening of welded areas, but also led to welding of interfacial regions which may have been in contact during rolling without achieving preliminary metallurgical bonding or which may have come into contact during annealing through thermal expansion of metals. Thus, the bonding strength was assisted by diffusion phenomena at high temperatures ($>850^{\circ}\text{C}$).

Finally, the growing porosities observed along the interface may also have led to reduction in bonding strength for the highest temperatures. These porosities may have been formed due to the Kirkendall effect [10] or due to the swelling of air bubbles trapped at the interface during rolling. This phenomenon is supposed to be a major reason for saturation of bonding strength beyond 1050°C . Besides, grain growth causing mechanical softening in welded areas can also have contributed to the bonding strength reduction and may have become critical compared to the beneficial effect of diffusion.

4. Conclusion

The effects of annealing treatments on the bonding strength of a bimetal strip consisting of two different iron-nickel alloys were investigated. The following conclusions were drawn from this work:

- As annealing temperature rose, the bonding strength was significantly improved, reaching its maximum at 1050°C .
- It was suggested that the increase of bonding strength, first promoted by grain growth from 650°C , was also assisted at higher temperatures by interfacial diffusion, which helped to improve the metallurgical bonding in welded areas and expand the region of welding.
- Despite the continuous increase of diffusion, the saturation of bonding strength beyond 1050°C is likely due to porosities along the interface and strong grain growth causing mechanical softening in welded areas.
- Nucleation of new grains was observed in deformation zones around fragments and interface regions. Further investigation is required to understand the influence of recrystallization and grain growth mechanisms on the increase of interfacial bonding.

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