Microstructural Characteristics of Accumulative Roll Bonded Ni-Al Based Metal Intermetallic Laminate Composite

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Abstract

The present investigation is an attempt to develop metal-intermetallic laminate composites based on Ni-Al system. In this study, Ni and Al foils (460 and 90 μm respectively) have been used for the development of Ni-Al laminate using accumulative roll bonding technique at 773 K. The laminate composites were then subjected to controlled annealing at 773 K, 823 K and 873 K for 1, 2 and 4 h to affect reaction at the Ni/Al interface leading to intermetallic compound formation. The accumulative roll bonded laminates showed good bonding of layers. Annealing treatment at 773 K led to slow formation of reaction product but maintained the interface integrity. Annealing at 873 K led to de-lamination of individual layers prior to the completion of reaction. A qualitative compositional analysis at the interfaces reflected the formation of Al-Ni copmpounds, and also a gradual compositional gradient across the interface. This process seems to be of promise so far as the continuous production of large scale metal-intermetallic laminate composites are concerned. A further study is underway to circumvent the delamination problems and property evaluation.

Keywords: Metal intermetallic composite, accumulative roll bonding, Al-Ni, Annealing

INTRODUCTION

Metal matrix composites have gained tremendous interest due to their unique combination of both mechanical and physical properties, such as high stiffness and high abrasion resistance. However, high strength metal reinforced with intermetallic compounds have attracted more emphasis compared to conventional ceramic particle reinforced metal matrix composites. The requirement of high strength at elevated temperatures has led to the development of metal-intermetallic laminate composites [1-2]. The combination of metal-intermetallic laminates (MILs) has been introduced to overcome the brittleness of monolithic intermetallics at ambient temperatures. These composites offer superior properties compared to constituent phases, for example, high bending strength coupled with low density and oxidation resistance [3-5]. Several MILs have been developed so far including Fe-Al [6-7], Ni-Al [1,5], Ti-Al [4,8], Cu-Al [9] etc. However, Ni-Al based laminate composites are considered to be economical and of paramount importance for high temperature structural and armour applications.

In general, these laminate composites are produced by employing constituent metallic foils stacked together, and affecting reaction between individual metallic layers under high pressure (15-50 MPa) and high temperature (1073-1373 K) [1, 4-5]. This technique utilizes high temperature and pressure so as to effect close contact between the individual layers and increased diffusion rate. The effect of pressure and temperature on the resulting composite, and the phases formed thereof at the interface have been widely studied. There are several variants of this route employed to reduce the cost of processing and as well as improve the properties of the composites. Recently, Mizuuchi et al. [1] have reported pulsed current hot pressing technique to realize such laminate composites, where pulsed current is used to effect local melting of the material and obtain pore free intermetallic layers. Ding et al. [10] showed the possibility of hot pressing followed by cold rolling and annealing treatment to produce Ni-Ti multilayers. However, these processing techniques do not seem to be economically viable routes when widespread industrial application is sought. We, therefore, attempted to employ accumulative roll bonding technique to first produce layered bonded composites of individual metallic layers, followed by controlled heat treatment, to realize metal intermetallic composite. The evolution of microstructural features of the composites have been reported and discussed.

EXPERIMENTAL DETAILS

The materials used in the present study were commercially available Aluminium foils (0.09×30×90 mm) and pure Ni sheets (0.46×30×90 mm). The individual Ni sheets (460 μm) and Al foils (90 μm) were wire-brushed to remove the surface oxide layers and cleaned with acetone so as to affect proper metallic contact due to compressive loading during rolling. Initially, four Aluminium foils were alternatively placed between five Nickel sheets. The edges of the so formed stack were trimmed. The stack was then fastened together by using thin steel wires (0.5 mm diameter) to prevent slipping of individual layers during the rolling process. Before the rolling experiment, the layered stack was soaked at 773 K for 5 min. It was then immediately rolled dry (the rolls were thoroughly cleaned to avoid slipping during roll pass) at a speed of 26 fpm. The diameter of rolls was 300 mm. The rolled sample was then cut into two halves and stacked again by placing an Aluminium foil of the same dimension, after proper surface cleaning. The whole process of rolling was repeated again. This implies that the Al layer used in the 1st pass decreases in thickness, while the Al layer used in 2nd pass remains as in the first pass. The details of the layer thickness and sample characteristics are shown in Tables 1 and 2.

Table 1 shows that a single pass deformation of around 50-55% in thickness gives rise to good bonding between Ni-Al layers. Table 2 shows that a cumulative strain for Ni is 0.93 and 1.79, respectively, for 1st and 2nd pass.. This is in contrast to the strain experienced by Al layer, which is 1.35 and 2.44 in the 1st and 2nd pass respectively.

The samples from both the 1st and 2nd pass laminates were taken for annealing treatment to affect the formation of intermetallic layers at Ni-Al interfaces by reactive diffusion. The samples were kept in a resistance heating furnace at different temperatures (773 K, 823 K and 873 K) for different time duration (1 h, 2 h and 4 h). Samples of suitable size from the 1st and 2nd pass were cut in the transverse direction from the center of each specimen. All samples were mounted in a hot mounting press using a conductive type mounting compound (Black Carbon). Mounted samples were then polished using standard metallographic technique. The polishing of the samples was carried out using 0.3 μm alumina and 0.05 μm diamond suspension. The samples were studied under scanning electron microscope (HITACHI S-3400N) attached with compositional line scan facility. Qualitative compositional variation was recorded across the Ni-Al interface.

Table 1 : Stack thickness and % reduction after first and second pass

Pass	**Layers**		**Stack thickness (mm)**		**% reduction**	**Bonding**
	Ni	**Al**	**Initial**	**Final**		
1st	5	4	2.66	1.19	55.26	Good
2nd	10	9	2.47	1.20	51.42	Good

Table 2 : True strain values of individual layers after first and second pass

Pass	**Thickness (μm)**				**True strain**		**Cumulative strain**	
	Ni initial	**Al initial**	**Ni final**	**Al final**	**Ni**	**Al**	**Ni**	**Al**
1st	460	100	206	30.99	0.93	1.35	0.93	1.35
2nd	206	30.99	97.8	12.09	0.86	1.08	1.79	2.44

RESULTS AND DISCUSSION

Microstructural characteristics

The procedure of manufacturing layered Ni-Al composites was successfully utilized in this study. Layered structure could be obtained with no trace of debonding at the Ni-Al interfaces. Roll bonding experiments were carried out at room temperature and 573 K, but it resulted in poor bonding at the interface. Whereas, a high temperature of 773 K led to a good bonding. However, at the temperature of 873 K, Al layer deformed extensively which was not required in this study. Therefore, 773 K was chosen as optimized temperature for further processing. Also, lower temperature of 773 K gave rise to limited effect on the material deterioration by surface oxidation that hampers good bonding. Figs. 1 (a-d) show the structure of the laminates in as rolled condition i.e. before annealing treatments. Figure 1a and 1b shows the structure after 1st and 2nd pass respectively. It is obvious from the figures that the Al layers could not remain equidistant, which is due to the fact that the rolling process may affect uneven deformation in the samples. The unevenness may also arise from the small variation in thickness during wire brushing. This part of the process is not controlled in the present study. This feature is not seen in the layered composites made by hot pressing of dissimilar metal foil stack, where no deformation is involved [5]. Figs. 1c and 1d shows high magnification micrographs, which reveals a good bonding between Ni and Al layers, both after the 1st and 2nd rolling passes, respectively. It is also worth mentioning that there is not any reaction product at the interface during soaking and rolling stages of processing.

Figs. 2 (a-f) show the microstructural features after annealing treatment at 773 K for different durations. These micrographs indicate a gradual growth of intermetallic layer at the Ni-Al interface. Figs. 2(a-c) are the micrograpahs of the laminate after 1st pass annealed for 1, 2 and 4 h respectively. It clearly indicates that initial reaction (i.e. after 1 h) does not take place throughout the interface. And also, there are some spikes or blurred interface indicating diffusion of Al at the interface. This may be due to the high dislocation density generated at the interface and dislocations act as easy path for diffusion. This is also manifested in the fact that the growth takes place from the interface into the Ni matrix (fig. 2a). As the time of annealing increases, more interfacial area is covered with the reaction product. Moreover, it is observed in fig. 2b that there are two layers growing at the interface having different compositions. After the completion of 4 h, almost whole interface is delineated with a reaction product. If we look at the growth morphology after 2nd pass, it is readily seen that most of the Al layer is transformed just after 1 h. After 2 h at 773 K, samples from 2nd pass almost transformed to intermetallic phase leaving a hairline Al layer. This enhancement in the transformation kinetics can be attributed to the high accumulative strain experienced by both the Ni and Al layers during rolling passes. This is in contrast to the MILs made by thin foils using hot pressing technique, where a high temperature of approximately 1273-1373 K is required. In the present investigation, the Al layer utilized is thicker than that employed in the former processing route [1, 2]. Fig. 2f shows that all the Al is consumed to produce intermetallic compound, whereas, leaving large amount of internal decohesion. The white particles have been identified as some compound of Ni and Al. This behaviour indicates that a restrictive force is required to avoid decohesion due to residual stresses developed at high temperature processing. The residual stress might have generated during cooling due to different thermal expansion coefficients of layers.

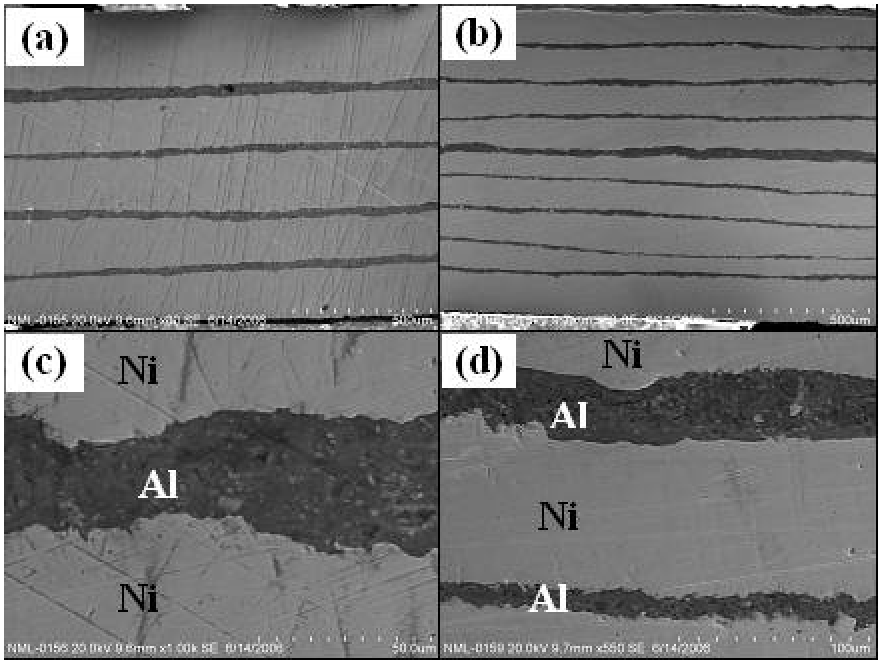

Figure 1: (a-b) layered structure after 1st and 2nd pass respectively (c-d) high magnification micrographs showing good bonding in after 1st and 2nd pass respectively.

The microstructural features of the samples annealed at higher temperatures are shown in figure 3. Figure 3a shows the two different Al layers (from the 1st as well as 2nd pass) indicating different extent of reaction at 773 K for 1 h, depending upon the strain value experience by individual layers. The middle layer, introduced in the second pass, experienced low strain compared to those came from the first pass. Therefore, the middle layer shows less amount of reaction products. Figure 3b shows that at 823 K (1st pass) and 4 h almost all the Al is consumed. At the same temperature (2nd pass), figs. 3c and 3d show micrographs after 2 and 4 h annealing, respectively, indicating no difference in the microstructural features. However, at this temperature, a small decohesion is developed at the interface to compensate for the volume change during tranformation. At 873 K, it is clearly exhibited in the fig. 3e that the problem of delamination arises due to a large difference in the thermal expansion coefficient of the two layers. It was observed that delamination occurs at a very early stage before the completion of reaction at the interface.

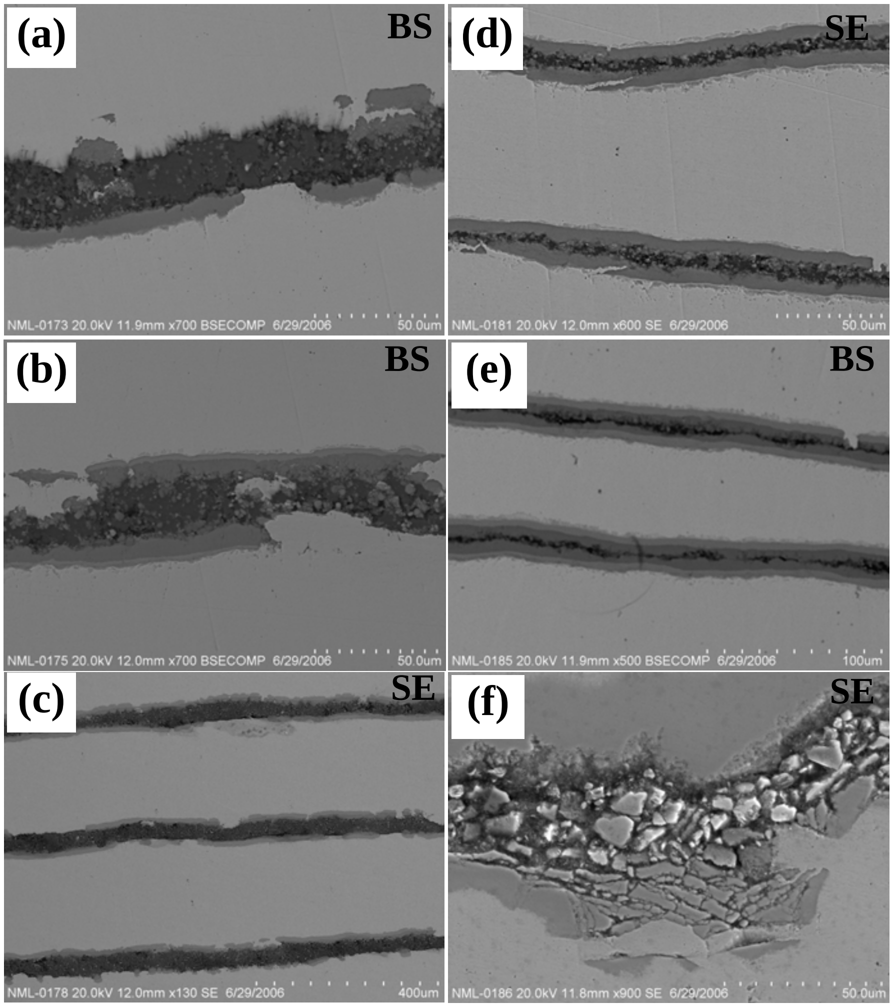

Figure 2: Micrographs of (a-c) 1st pass laminate annealed for 1, 2 and 4 h respectively (d-f) 2nd pass laminate annealed for 1, 2 and 4 h respectively.

Compositional variation

The compositional variation at and near the interface has been characterized by line scan. Fig. 4a shows the qualitative compositional profile across the interface for 1st pass, annealed at 773 K for 1 h. It seems that Al diffuses to Ni matrix leaving some areas unaffected. This is due to the fact that an increased dislocation density have been introduced during deformation and these dislocations act as shortest path for the diffusion of Al. Otherwise, the Al and Ni layers are clearly distinguishable from the line plot. The presence of Ni is negligible in the Al layer. A gradual decrease of the Ni concentration at the interface indicates that there is no sharp boundary between Al and Ni layers. After 4 h of annealing at the above condition, a continuous layer forms at the interface. However, a backscattered image (as shown in fig. 4b) shows two distinct layers indicating the presence of Ni rich and Ni lean layers. As discussed by Mizuuchi et al. [1], the compositional gradient may lead to a better bonding between Ni and Al layers and also the Ni and Ni aluminide in the later stages of annealing. If we compare figs. 4a and 4b, it is clearly visible that in the latter case the Al layer profile does not change sharply as seen in the former. This indicates that Ni diffuses to Al layer with time. However, at higher temperature of 823 K, the presence of two layers is not seen.

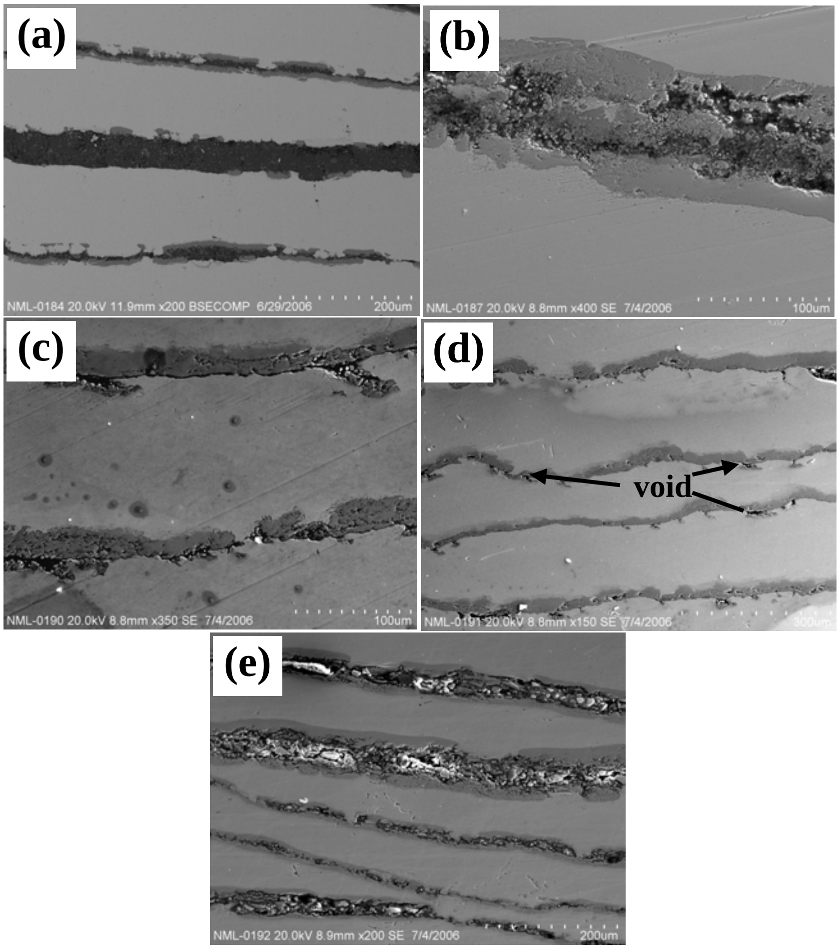

Figure 3: Micrographs of the annealed samples in the condition (a) 1st pass, 773 K, 1h (b) 1st pass, 823 K, 4 h (c) 2nd pass, 823 K, 2 h (d) 2nd pass, 823 K, 4 h and (e) 2nd pass, 873 K, 2 h.

Fig. 4c shows the line profile as well as the micrographs of the sample (1st pass, 823 K, 4h) showing almost full consumption of Al. As can be seen from the line profile, only about 10 μm thick Al remains unreacted where Ni has not gone into solution. If we look at the plot within the two dotted line (figure 4c), it is obvious that the loss of Al at one end is compensated on the other. Whereas, the compositional profile outside the dotted line are almost symmetrical. Fig. 4d shows the line scan for the sample (2nd pass, 550 °C, 4 h) indicating full transformation of Al into Ni-Al compound.

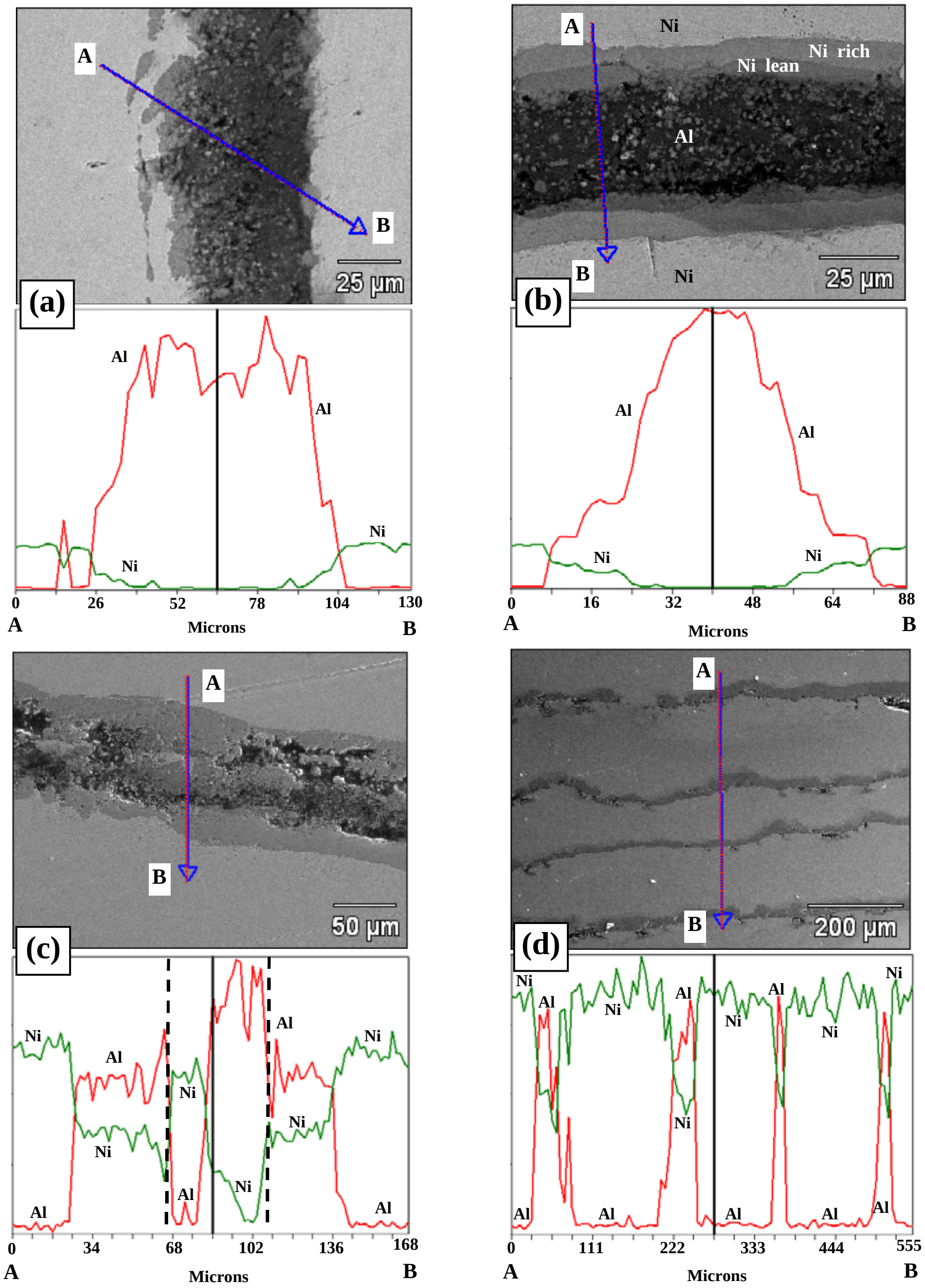

Figure 4: Compositional line scan across the interfaces for the conditions (a) 1st pass, 773 K, 1 h (b) 1st pass, 773 K, 4 h (c) 1st pass, 823 K, 4 h (d) 2nd pass, 873 K, 4 h.

A high temperature processing above 973 K [1] leads to melting of Al layer and therefore a solid-liquid reaction couple is formed. The solid state reaction in this study, requires high activation energy for the reaction to take place. In general, Ni and Al react at the interface giving rise to $NiAl_3$. The binary equilibrium phase diagram [13] indicates that the first phase to form in solid sate Al-Ni solution is $NiAl_3$. Further, with increased diffusion time $NiAl_3$ transforms to

Ni_2Al_3 [5,11,12]. However, transformation to NiAl and Ni_3Al requires a high annealing temperature and longer duration. Therefore, it is expected that the formation of latter two phases would not have taken place in the present case. The presence of two distinct layers in figs. 3 (c and e) may be construed to be $NiAl_3$ (inner layer) and Ni_2Al_3 (outer layer) based on the above discussion. However, as indicated in fig 4d (2nd pass, 2 h, 823 K), the second layer is absent. The full transformation of inner layer ($NiAl_3$) to Ni_2Al_3 may be the reason for the absence of two layers.

The present study, therefore, dealt with the development of Ni based Ni-aluminide laminate composites. However, the general observation indicated that the annealing treatment after repetitive roll bonding led to formation of voids and also de-lamination at higher temperatures. Generally, such reaction synthesis is carried out at higher temperatures. This problem can be avoided by applying a positive pressure during annealing treatment. However, in the present study, the effect of high strain accumulation during rolling was made use of to affect complete reaction at lower temperatures and short annealing duration. Though the grain refinement study has not been carried out in this investigation, it is well known that this kind of high strain processing lead to grain refinement of the matrix phase giving rise to improved properties. The qualitative compositional analysis indicated that the diffusion across the interface is a function of temperature as well as time of annealing.

CONCLUSIONS

The present investigation is an initial attempt to develop economically viable production route for the metal-intermetallic laminate composites. Thick foils have been used for making laminates of Ni-Al by accumulative roll bonding followed by controlled annealing at 773 K, 823 K and 873 K to affect consumption of Al foils to form intermetallic compound. It was observed that the reaction after 2nd pass of roll bonding was faster compared to that after the 1st pass. The high strain accumulation during rolling has been concluded to be effective in faster reaction at the interface. Low temperature of 773 K and longer duration of annealing is more effective to avoid de-lamination of individual layers and also a good adherence of intermetallic compounds.

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