Effect of silicon, manganese and nickel present in iron on the intermetallic growth at iron - aluminum alloy interface

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ABSTRACT

Recent interest in joining aluminum to steel necessitates a critical understanding of growth kinetics of various intermetallic phases that form across the mixed material interface. In order to understand the individual effect of ternary alloying elements present on iron side, growth kinetics of intermetallic layers formed at the interface between iron and aluminum alloy 6061 were studied by dipping pure iron and binary Fe-X alloys in the melt of AA6061, where X was Mn, Ni and Si. The immersion experiments were performed at 750 °C for various time durations from 90 s to 2400 s. The intermetallic layers formed at the interface were Fe_2Al_5 and $FeAl_3$ in all cases. It was found that the presence of all the three ternary alloying elements in iron reduced the growth of intermetallic layer. Silicon was found to be the best for reducing the growth kinetics of the intermetallics followed by Ni and Mn in that order. The growth of Fe_2Al_5 layer was found to be diffusion controlled. Thickness of $FeAl_3$ layer was found to increase with time at the beginning and to decrease with time at later stages. This behavior of $FeAl_3$ was attributed to the two competitive processes that are simultaneously occurring at the $FeAl_3$/AA6061 interface during liquid-solid diffusion viz. dissolution and formation of the $FeAl_3$. The effect of the alloying elements on the growth kinetics of the intermetallic layer is explained based on velocity of interface in terms of inter-diffusion fluxes and concentration gradients across the interface.

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1. Introduction

Aluminum has been projected as the most promising light-weight material for replacing conventional steels for structural applications in automobiles [1—4]. However, the constraints on cost and properties of aluminum alloys allow the designers to replace only those car body parts that do not require very high strength such as hood, trunk, door and roof [5]. This necessitates the development of hybrid-material technologies that use hybrid structures of steels and aluminum alloys and thus, joining of aluminum to steel has become essential in automotive industry [3,4,6]. Various methods can be used for metallurgical joining of aluminum to steel including fusion welding, explosive welding, laser welding, friction stir welding and diffusion bonding [7—11]. Among these, fusion welding processes such as resistance spot

welding are the most economical and most commonly used methods to join aluminum and steel [3,6,12]. Producing intermetallic-free joints is the most critical challenge in fusion welding of aluminum alloys to steels [7,13—18].

Fig. 1 presents the binary phase diagram of Fe-Al system [18,19]. The solubility of iron in solid aluminum is very low (<0.1 wt % at 600 °C). On the other hand, aluminum shows a large solubility in α-iron (space group $Im\overline{3}m$, 229, Pearson symbol *cI2,* structure type W) i.e. up to 28 wt % of aluminum at 1310 °C.

Six types of intermetallic phases can be seen on the Fe-Al phase diagram viz. Fe_3Al, $FeAl$, $FeAl_2$, Fe_2Al_3 (denoted as ε in Fig. 1), Fe_2Al_5 and $FeAl_3$. The last two phases have also been designated as $FeAl_{2.8}$ (the space group *Cmcm*, 63; the Pearson symbol oS24; the structural type $FeAl_{2.8}$) and Fe_4Al_{13} (the space group C2/m1, 12; the Pearson symbol mS102; the structural type Fe_4Al_{13}) respectively [20—22]. However, the two phases will be denoted as Fe_2Al_5 and $FeAl_3$ for the rest of this manuscript. It should be noted that not all intermetallic phases form in the weld joints. The most commonly observed intermetallic phases in welded joints of steel and aluminum alloys are Fe_2Al_5 and $FeAl_3$ [13—18]. These phases are
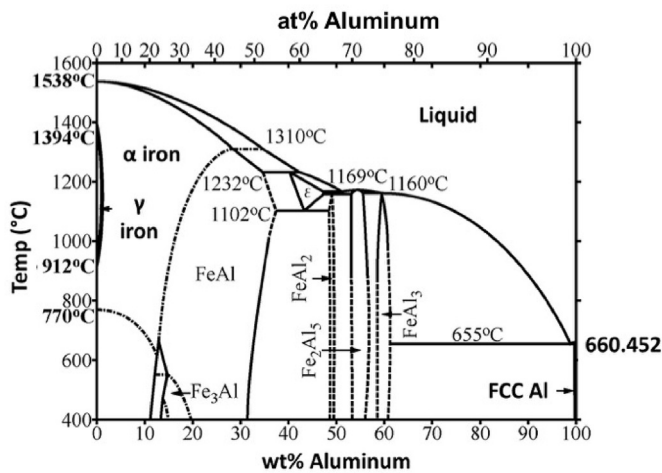

Fig. 1. Fe-Al phase diagram [18,19].

brittle and their presence at the interface deteriorates the joint strength. The thickness and morphology of the intermetallic layer plays a crucial role in determining the joint strength. Various studies have shown that reducing the intermetallic layer thickness increases the fracture toughness and shear strength of the weld joints [7,13,23–25]. It should be noted that the actual value of maximum tolerable intermetallic thickness would depend upon the design of the component and the service conditions. The welding process has to be designed so as to keep the intermetallic layer thickness below the maximum allowed by the design. Thus, it is very important to know the factors that govern the growth of intermetallic layer during joining of aluminum and steel, so that thickness can be controlled for obtaining the desired joint strength. Several authors studied the growth kinetics of intermetallic layer formation by controlled dipping experiments of pure iron or steel in aluminum [26–32] and the growth has been observed to be diffusion controlled in most cases [29–33]. To minimize the growth of intermetallic layer during fusion joining of aluminum to steel, we should maintain low temperature and reduce the time of operation. But time and temperature reduction have certain limitations during welding and cannot be reduced beyond a certain limit. So, it is necessary to also be able to influence other parameters to control the growth of intermetallic layer. Alloying elements, when present either in steel or in aluminum, can affect the diffusivity of Fe and Al and in turn, can affect the formation and growth of intermetallic layer. Akdeniz and Mekhrabov [34] have proposed a model for predicting the effect of substitutional impurities in iron on the evolution of Fe-Al diffusion layer by calculating the pairwise interatomic interaction potentials, which were used to predict the activity of Al atoms in α-Fe$_{0.95}$ (Al$_{1-n}$X$_n$)$_{0.05}$ where X is an alloying element like Si, Mn, Ni, Cr etc. Their models predicted that Si, Ti, Cu and Mg retarded the growth of intermetallic layer as these elements reduced the activity of Al in steel, whereas Zn, Mn and Ni accelerated the growth of intermetallic layer because they increased the activity of Al in steel. However, the model proposed by them is based solely on thermodynamic aspects and no consideration for kinetic aspects such as diffusivities is included. In most of the studies reported in the literature so far, the effect of addition of alloying elements on aluminum side has been studied [35–41]. It was found that addition of Si in Al melt reduced the thickness of intermetallic layer as compared to pure Al melt [36–38]. Mg in Al melt also reduced the growth of intermetallic layer [39]. The effect of Cu, Sr and Ti was also studied and these elements also led to reduction in thickness of intermetallic layer

[40,41]. Very few studies have reported effect of alloying elements present in steel on the growth kinetics of intermetallic layer at the interface. Su et al. [42] found that the activation energy for the intermetallic growth reduced when Fe-8Al-30Mn-0.8C (wt%) was dipped in pure aluminum bath. Hawang et al. [43] studied the effect of carbon content of steel on formation of intermetallic layer and dissolution of steel in molten Al at 660 °C. It was found that the dissolution rate and growth kinetics decreased with an increase in carbon content. The decrease in thickness of intermetallic layer was attributed to the reduction in diffusion rate of Fe and Al through the layer.

In general, the effect of various alloying elements present in Al-alloys on intermetallic layer has been reported widely. However, the effect of individual alloying elements present in steel on the formation and growth of intermetallic layers at the interface between iron and aluminum alloys has been given almost negligible attention. In the present work, the effect of small amount of alloy addition (Mn, Ni and Si) to iron on intermetallic layer growth at the iron/aluminum-alloy interface was investigated. The three elements chosen are the common alloying elements present in mild steels, which are most often encountered in steel-aluminum hybrid structures of auto body parts. The explanation for the effect of alloying elements on the growth kinetics of intermetallic is also provided based on the interdiffusion fluxes and concentrations of the elements developed at the interface.

2. Experimental work

Pure iron and three different alloys with nominal composition of Fe-2 at% X (where X = Mn, Ni or Si) were used for the present investigations. The alloys were prepared in a vacuum arc melting furnace. The pure elements used for preparation of the alloys viz. 99.98% Fe, 99.95% Mn, 99.95% Ni and 99.99% Si (all purities in wt%) were obtained from Alpha Aesers. The arc melting furnace chamber was evacuated to approximately 10^{-5} torr and purged with Argon two times and the final melting was carried out in a low pressure Argon atmosphere. The alloy buttons were re-melted four times, each time flipping the button upside down. This was done to ensure uniform mixing of alloying elements and to avoid any major segregations. Each alloy button weighed approximately 15 g. The alloy buttons were then cut using slow diamond-wheel cutter and three discs were cut from each alloy. Each sample was then cold rolled (~20%) to provide driving force for recrystallization during subsequent homogenization treatment. The rolled alloys were then sealed in a quartz tube, which was evacuated and filled with argon and homogenized at 1100 °C ± 5 °C for 48 h in a muffle furnace, followed by air cooling. After homogenization, all three discs from each alloy were further cut into 6 samples of dimensions approximately $5 \times 5 \times 3$ mm^3. All the samples were then metallographically polished followed by ultra-sonic cleaning in methanol for 15–20 min. After cleaning, the liquid-solid diffusion couples were formed by immersion of the pure iron or the Fe-X alloy in liquid aluminum alloy AA6061. For the immersion experiments, firstly AA6061 samples of dimensions $10 \times 2 \times 1$ cm^3 were polished using emery paper 180 and cleaned using ultra-sonic cleaner. Table 1 presents the chemical composition of the binary Fe-X alloys measured with EDS (average of twenty points each). The composition of AA6061 as measured using optical emission spectroscopy

Table 1
Chemical compositions of Fe-X alloys.

Conc. of X in Fe-X	Fe-Mn	Fe-Si	Fe-Ni
at %	2.0 ± 0.1	2.6 ± 0.4	1.9 ± 0.1