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Effect of silicon on the microstructure and growth kinetics of intermetallic phases formed during hot-dip aluminizing of ferritic steel

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Highlights

- Microstructure of hot-dip aluminized steel with rising Si content was investigated.
- Si addition caused a reduction in η phase thickness and formation of ternary phases.
- In the θ and η phase were respectively micro twins and a wavy structure observed.
- Growth kinetics were investigated by dipping at various temperatures and times.
- Activation energy for the η phase decreased with rising Si content in the Al bath.

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Introduction

Dissimilar joining of aluminium (Al) and iron (Fe) based materials is of significant importance for engineering design. For structural applications the lower mass density of Al alloys is brought together with the higher strength and stiffness of Fe based steels, for example to reduce the weight of transportation systems in order to improve their performance and meet ever more stringent environmental legislation constraints [1]. Another important application is combining the superior corrosion resistance of Al – especially at elevated temperatures [2] – to the high strength of the steel substrates. This is widely realized by coating techniques, of which the most versatile, economic and commonly applied technique is dipping the steel substrate in a molten Al bath, referred to as hot-dip aluminizing (HDA) [3]. The main challenge in HDA, and dissimilar Al-steel joining in general, is the formation of inherently brittle intermetallic phases (IMPs) at the interface between Al alloys and steel, as they are the main cause of cracking and spalling of the coating during forming operations [4]. In the case of pure Al coatings, the intermetallic seam consists of two different IMPs, namely the θ phase ($\text{Fe}_4\text{Al}_{13}$) adjacent to Al and the η phase (Fe_2Al_5) in between θ phase and steel. The η phase has been found to be the rate controlling component, rendering it of critical importance for the joint performance [5], [6], [7], [8], [9].

Heumann and Dittrich [10] were among the first to report on the parabolic growth kinetics of the intermetallic layer, assuming it to solely consist of η phase. Denner and Jones [11] also indicated a linear dependence for the $\sqrt{t}$ kinetics, but noted deviations from this relationship at longer reaction times. Eggeler et al. [12] detected the presence of a thin θ phase layer in between the Al and η layer. Fe saturation of the Al bath were shown to shift the interface between Al and intermetallic layer more outward compared to the pure Al, and the activation energies ΔH of for the growth of the reaction layer were about 155kJmol^{-1} for pure Al and 104kJmol^{-1} for Fe-enriched Al melts, respectively [7]. When maximum and not mean thicknesses were investigated, the values of the activation energies were about 20kJmol^{-1} lower.

The main strategy to reduce the thickness of the IMP layers is the addition of silicon (Si) to the Al bath as it – additional to lowering the melting temperature and viscosity of the bath – suppresses the growth of the η phase [4], [13], [14]. Numerous investigations have been conducted on the underlying microstructural phenomena, but the mechanisms behind this growth retardation are not yet fully understood [13], [15], [16], [17], [18], [19], [20], [21]. Nicholls [16] proposed that Si atoms might interact with the open crystallographic arrangement of the η phase, and consequently decrease the growth kinetics. Alternative hypotheses suggest a reduced activity of Al [15], [21], the formation of a ternary phase acting as a diffusion barrier [20], an influence on the activity coefficient of the Si, or a reduction of the activation energy for the η phase formation [13].

[18], [19]. Recently the presence of Si-enrichments at grain and phase boundaries of the η and θ phase could be detected by atom probe tomography investigations, these enrichments might also affect the interdiffusion processes and thus growth kinetics of the IMP seam [22]. Apart from the reduction in η phase thickness, Si additions also cause a change in morphology of the interface from finger-like to a more planar structure, and the formation of ternary phases [18], [23]. A reasonable understanding of the different types of Al-Fe-Si ternary phases is reflected by literature, but the phases have quite large and closely positioned homogeneity ranges. Despite some variation in nomenclature they are generally quoted as τ_1 to τ_{10} [18], [23]. Additionally the influence of the substrate must not be underestimated, the addition of Si to the steel [24] or whether it is austenitic can have an effect as well [25], [26].

It thus becomes clear that the Si-induced growth suppression of IMPs in dissimilar joining of Al and steel is a complex scenario of influences and consequences. Comparing and evaluating literature data in this field, however, is difficult since significantly different dipping temperatures and times, bath and substrate compositions, as well as procedures for calculating growth kinetics have been used in the different investigations. Consequently, a comprehensive and systematic study of the influence of varying Si concentrations and interdiffusion conditions on the build-up and growth kinetics of the IMP seam is of high interest. Therefore, this work aims to contribute to the understanding on the role of Si in the growth kinetics as well as the microscopic characteristics of the different intermetallic phases formed. After hot-dipping in Al baths with systematically varied Si contents, temperatures and dipping time, the thickness and morphology are analysed to elucidate the microstructural parameters which control the reaction kinetics of the intermetallic phase formation.

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Materials and methods

Hot-dipping experiments were performed with a Rhesca hot-dip simulator under an atmosphere of N_2 -5% H_2 to prevent oxidation of the steel. DC06 low carbon European grade steel sheets (Fe-0.01C-0.015Si-0.1Mn-0.01Cr-0.018Ni-0.05Al-0.01Cu-0.04Ti) of 150 by 100mm and a thickness of around 1 mm were dipped in aluminium baths containing a

Si contents of 0.3, 1, 3, 5 and 10wt%, respectively. A content of up to 0.3wt% Si in such a bath is currently considered in industrial applications as “pure” ...

Microstructure

Representative EBSD results are shown in Fig. 1 as phase maps of the technically pure Al, Al-3, Al-5 and Al-10wt% Si experiments dipped for 60s at temperatures of about 685°C. Blue represents Al, yellow the θ phase, green the η phase and red Fe α (i.e. the steel substrate). The ternary phase τ_5 which is formed at higher Si contents is shown in light blue. Image quality data are superimposed on the phase maps to indicate grain boundaries. The η phase is thicker than the θ phase and contains the ...

Discussion

For pure Al we observe an activation energy for η layer growth of around 224kJmol⁻¹ which is slightly higher but still in good agreement with previously reported values of 190kJmol⁻¹ [18] and 207kJmol⁻¹ [19]. From Table 1 it can be observed that with increasing Si content the activation energy decreases. This dependence was also reported previously in literature, but only calculated from literature data [18] or for baths up to 3wt% Si [19]. A value for the Al-10wt% Si case was not reported ...

Conclusion

The influence of Si addition to the Al bath on the intermetallic phase formation during HDA was investigated for temperatures between 670 and 725°C and various dipping times. From the obtained results following conclusions can be drawn:

- 1) New insights into the microstructure and growth kinetics of the intermetallic phases formed by hot-dip aluminizing could be obtained with EBSD and SEM investigations. ...
- 2) For lower Si contents, the IMP seam consisted of the major η phase and a thin θ phase. The η ...

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First page preview

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1. Introduction

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...It was clear that the intermetallic layer with a tongue-like structure was rather flat compared with that of the soft steel. This phenomenon may be due to the Si in the cast iron [28]. It is generally considered that the addition of Si prevents the diffusion of Al and reduces the growth rate of Fe_2Al_5

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...The slurry pre-coating process is intended to increase the concentration of aluminum atoms on the substrate surface, which contributes to reducing the temperature required for aluminizing. The aluminizing temperature in this process was 680 °C, much lower than the traditional powder embedded aluminizing temperature which is in the range of 900–1150 °C [8,34], but similar to the HDA temperature which is generally 700–750 °C [5,22,35–37]. This process innovatively combines dipping aluminizing at room temperature with powder embedded aluminizing, which not only circumvents the voids that are easily produced in hot-dipping aluminizing, but also reduces the temperature of embedded aluminizing....

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