Evolution of Al-Si Coating Microstructure during Heat-Treatment of Usibor® 1500

I Yakubtsov* and R Sohmshetty

Ford Motor Company, Dearborn, Michigan, 48124, USA

*E-mail: iyakubts@ford.com

Abstract. Microstructural evolution of Al-10wt.%Si-(2-3)wt.%Fe coating during heat-treatment processing of the press hardenable steel Usibor® 1500 was studied experimentally. Two coating thicknesses, AS150 (60-100 g/m²) and AS80 (35-65 g/m²), were subjected to the same heat-treatment schedule. The interrupted experiments were performed at heating and soaking stages in order to track the microstructural evolution. The effects of heating rates (and steel substrate thickness) on evolution of the coating were researched as well. Furthermore, the experimental results of coating development were then compared with the results of analytical modelling.

1. Introduction

The most effective way to produce automotive parts (for example A-pillar, B-pillar, bumper, impact beam, roof rail and tunnel) from ultra-high strength steels (UHSS) is hot stamping (HS) or hot press forming (HPF). Coated steel sheets are used in order to prevent surface oxidation and decarburization during HS/HPF processing, and to improve corrosion resistance of the produced parts. Currently, Al- and Zn-based metallic coatings are extensively used for HS/HPF production in the automotive industry. Aluminized coating of type 1 consists of Al - (7-11) wt.%Si alloys which are close to the eutectic composition. The type 2 of aluminized coating is pure Al. Silicon content in Al - Si coatings strongly affects the formation kinetics of Fe- and Al-rich intermetallic phases throughout HS/HPF processing [1-10]. The mechanical and electrical properties of Fe- and Al-rich intermetallics are different [11-13], and their compositions have an impact on the overall properties of the Al - Si coating. Therefore, the properties (weldability, corrosion and paintability) of coated sheet steels depend on the microstructure and the volume fractions of intermetallics in the formed Al - Si coating after HS/HPF processing. According to the experimental observations, melting of the Al - Si coating occurs at about 580 °C and its solidification follows during a heating stage of HS/HPF [14]. The melted coating interacts with conveying rollers in the furnace during the heating stage of HS/HPF of production lines and causes a certain degree of damage to the rollers, and the damage influences the maintenance cost and service life of the rollers. The purpose of the current research is to explore the effects of HS/HPF parameters (heating rate, soaking temperature and dwell time) on microstructural evolution in the Al-Si coating of Usibor® 1500 at two different coating weights: AS150 (60-100 g/m²) and AS80 (35-65 g/m²).
2. Materials and Experimental Procedure
The material under investigation in this study is Usibor® 1500 with coating AS80 and AS150. Two sheet thicknesses, 1 mm and 1.8 mm, were used in the experiments. The samples of size 25.6 mm x 25.6 mm from each sheet were heat treated in the furnace in an air environment. The thermo-schedule is presented in Figure 1. The main parameters of the schedule are in Table 1. The heating rates were estimated in the temperature range between 25 °C and 870 °C. A K-type thermocouple with a diameter of 0.5 mm was positioned in a hole drilled in the side of the sheet, normal to the sheet thickness. This measurement set-up enabled observation of the temperature profile inside of the steel substrate. The soaking (austenization) temperature, $T_S$, was 900 °C. The dwell (furnace) time, $t_D$, was intentionally divided into the heating time, $t_H$, and soaking time, $t_S$, for the purpose of performing the interrupted heat-treatments at the heating and soaking stages. The interrupted heat-treatments were performed at each stage by taking the samples out from the furnace and quenching them in water at ambient temperature. Microstructure observations of the samples were carried out by optical microscopy. Coating micro-hardness measurements were estimated using the loads of 5 g and 25 g.

3. Results
3.1 Experimental Observations
Examples of the microstructural evolution at the heating stage are presented in Figure 2. The microstructural observations of as-received (raw) AS80 and AS150 materials have shown that the coating consists of an intermetallic layer of 4-6 µm thickness at the interface between the steel substrate (pearlite and ferrite) and the outer layer (Figure 2a). The outer layer is the metallic alloy Al – 10 wt.%Si - (2-3) wt.%Fe. The intermetallic layer in thin sheets (1 mm) is slightly thinner (1-2µm) compared to thicker sheets (1.8 mm). This can be explained by a slower cooling rate for the thicker steel sheet after its hot-dip aluminization at 650 °C in production conditions. In this situation, the diffusion process at high temperatures and longer times during the cooling provides the additional growth of the intermetallic layer. The subsequent application of air knives during the process enables the production of either AS80 or AS150 coating thickness at industrial scale. The intermetallic layers (or inter-diffusion
Figure 2. Heating stage, AS80, 1 mm: (a) as-received; (b) 650°C; (c) 750°C; (d) 900°C.

layer, IDL) in the as-received AS80 and AS150 coatings are very similar from a metallurgical point of view.

The examples of microstructural evolution of the coating in AS80 through the heating stage are presented in Figure 2. The melting temperature of the metallic alloy in outer coating layer is about 580°C to 590°C. Figure 2b shows newly formed areas of intermetallic due to solidification of the Aluminum liquid at 650°C. A similar behavior has been observed up until 850°C, at which point newly formed intermetallic and Aluminum liquids are each present simultaneously in both AS80 and AS150. Subsequent heating to 900°C provides complete solidification in every coating except AS150 in 1 mm sheet, where Aluminum liquid still exists in the outer layer.

The development of the coating at a soaking temperature of 900°C is presented in Figure 3. The overall microstructure of AS80 in both sheet thicknesses can be described by the formation of a continuous (or unbroken) four-layer microstructure: \( L_1, L_2, L_3 \) and \( L_4 \). \( L_1 \) is the layer at the interface between the steel and the coating, and \( L_4 \) is the outermost layer of the coating, with \( L_2 \) and \( L_3 \) being positioned between them. An increase in the soaking time leads to a broadening of \( L_1 \) and \( L_3 \), and shrinking of \( L_2 \) and \( L_4 \), respectively. AS150 shows a similar development pattern, except that \( L_3 \) is discontinuous (broken) as compared to AS80. Additionally, the middle areas of \( L_1 \) exhibited some formed voids due to Kirkendall effect. The formed voids in \( L_1 \) is a result of higher diffusivity for Fe in Al-rich intermetallic compared to Al diffusivity in austenite [3]. Both the size and the density of the voids increased with soaking time in each coating. Additional void formation with soaking time was observed in \( L_4 \) close to the outer surface in each coating.

Micro-hardness measurements have shown that the coating formed at 900 °C immediately after the heating stage is very brittle and its micro-hardness value is very high (about 1100 HVN). Indentation using a 25g load formed large cracks in the coating, whereas indentation using a 5g load created and indent successfully and without a crack in that coating (Figure 4).
Figure 3. Soaking stage at 900 °C, 1 mm: (a) AS80, 3 min; (b) AS80, 10 min; (c) AS150, 3 min; (d) AS150, 10 min.

Figure 4. Micro-hardness in AS80 coating after heating to 900°C followed by water quenching. Locations 1, 2, and 3 are indentations at 5 g, 25 g, and 25 g loading, respectively.

3.2 Modelling of Coating Growth Kinetics
Based on the experimental observations and the literature data, a schematic of the coating growth process is presented in Figure 5. After the heating stage, Al-rich intermetallics are formed with a sharp boundary between the steel and the coating (Figure 5a). At this condition, there is no inter-diffusion...
layer (IDL) and there are no voids. The subsequent soaking stage enables the formation of a four-layer microstructure in the overall coating, where \( L1 \) consists of two sub-layers: \( L11 \) at the coating/steel interface; and \( L12 \) at the \( L1/L2 \) interface (Figure 5b). The formations of \( L11 \) and \( L12 \) are due to the inter-diffusion of Al atoms from the coating into the steel, and of Fe atoms from the steel into the coating, respectively (Figures 5a and 5b). \( L11 \) is made of the \( \alpha \)-Fe phase (or Fe-Al ferrite,) and \( L12 \) is the Fe-rich intermetallic FeAl.

The model presented here covers the growth kinetics for the overall coating thickness (CT), as well as the \( L1 \) (or IDL) layer, and the equations for both are based on application of the diffusion principles which govern phase formation in solids. In this case, the growth of each phase can be described by the parabolic dependence:

\[
X (L11 \text{ or } L12) = k t^{1/2}
\]

\( X \) is a layer thickness; \( t \) is soaking time; \( k \) is a growth rate (\( \mu m/s^{1/2} \)) and \( k = k_o \exp(-Q/RT) \); \( k_o \) is a constant; \( Q \) is apparent activation energy; \( T \) is soaking temperature in Kelvin; \( R \) is a gas constant, and \( R = 8.31 J/(mole \times K) \).

The growth rate is obtained separately for \( L11 \) (\( \alpha \)-Fe) and \( L12 \) (FeAl intermetallic). Their total thickness along with the soaking time determines \( L1 \):

\[
L1 = L11 + L12 \quad (2)
\]

The growth rate and activation energy of \( L11 \) (\( \alpha \)-Fe) were obtained from experimental data in the literature at different soaking times and soaking temperatures (920°C, 950°C and 1000°C) for the AS150 coating as applied to Usibor 1500 steel: \( k (L11) = 0.37 \mu m/s^{1/2} \) at 900°C; \( Q (L11) = 182 kJ/mol \) [15, 16]. Using our experimental data for \( L1 \) (experimental) vs. \( t \) as well as the literature data for \( L11 \) vs. \( t \), we obtained \( k (L12) \) at 900°C:

\[
L12 = L1(experimental) - L11 = k(L12)t^{1/2} \quad (3)
\]

It was determined that \( k (L12) = k (FeAl) = 0.33 \mu m/s^{1/2} \) at 900°C.

The value reported in the literature for iron diffusivity, \( D(Fe) \), in the FeAl intermetallic is \( D(Fe) = 2.13 \times 10^{-14} m^2/s \) at 900°C; \( Q (FeAl) = 250 kJ/mol \) [17].

The value of \( Q_G (FeAl) = 250 kJ/mol \) can be used for calculation of the FeAl intermetallic growth rate at other temperatures according to \( k = k_o \exp(-Q_G(FeAl)/RT) \), because the growth of the FeAl intermetallic is governed by the diffusion of Fe atoms.

**Figure 5.** Schematic of coating:(a) before soaking, \( t_S = 0 \); (b) after soaking, \( t_S > 0 \).
The overall coating thickness CT is obtained by the following equation:

\[ CT(t) = CT(t = 0) + L11(t) \] (4)

where \( CT(t=0) \) is the coating thickness in the as-received condition, and \( t \) is soaking time.

The diffusion path of Fe atoms in the FeAl intermetallic is also estimated according to the expression:

\[ X(Fe) = 2(D(Fe)t^{1/2}) \] (5)

where \( D(Fe) \) is diffusivity of Fe in FeAl intermetallic. The combined results of modelling and experimental data are presented in Figure 6. The values of \( L12 \) and \( X(Fe) \) are very close. It confirms that FeAl intermetallic growth is governed by Fe diffusion in that particular intermetallic phase. The deviation of experimental and calculated data for CT in AS150 (1 mm) is most likely associated with the variation of coating thicknesses in the as-received condition which was observed through microstructural evaluation (Figure 6b).

**Figure 6.** Kinetics at 900°C:(a) \( L11 \) (1), \( L12 \) (2), \( L1 \) (3), \( X(Fe) \) (4); (b) estimated and experimental CT for AS80 and AS150 for 1 mm and 1.8 mm sheets.

The growth kinetics of \( L1 \) is lower at short dwell times (up to 220 seconds) compared to longer dwell times at 930°C (Figure 7). According to the literature data, there is an incubation time of up to 60 seconds.
seconds for the formation of ferrite (α-Fe) in medium carbon (0.1 to 0.5 wt% C) steels [18]. The incubation time depends on the temperature of ferrite formation and the chemistry (mostly carbon content) of the steel. In such a case, \( L1I \) is not yet formed and \( L1 = L12 \). Therefore, the calculation of \( L1 \) was performed for short dwell times (up to 220 seconds) at the condition \( L1 = L12 (L1I = 0) \) and for long dwell times according to equation (2) (Figure 7).

4. Conclusions
1. For the same hot stamping process conditions, the amount of liquid Al formed varies in the temperature range between 600°C to 870°C:
   - The liquid Al content is higher for thin sheets compared to thick sheets.
   - The liquid Al content is higher for AS150 compared to AS80.
2. The Al-rich intermetallics (\( \text{Al}_5\text{Fe}_2 / \text{Al}_3\text{Fe} \)) formed in the coating just after solidification are very brittle. Further soaking leads to formation of a more ductile intermetallic phase (\( \text{FeAl} \)) which reduces the amount of cracks formed during hot pressing and decreases potential porosity.
3. A four-layer structure of the coating (\( L1, L2, L3 \) and \( L4 \)) is formed during the first several minutes (about 1-3 min) of soaking time. All 4 layers are unbroken in AS80, whereas only \( L3 \) is broken in AS150.
4. The interdiffusion layer consists of two sub-layers of \( \alpha\text{-Fe} (L1I) \) and Fe-rich intermetallic (\( L12 \)), and vacancies/voids located in the \( \alpha\text{-Fe} \) due to Kirkendall effect. The growth kinetics of each sub-layer can be described by the parabolic dependence of its thickness vs. the square root of soaking time.
5. There is an incubation time for the formation of \( \alpha\text{-Fe} \). The formation of Fe-rich intermetallics is governed by Fe diffusion in the intermetallic coating.

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