Fusion zone microstructure evolution of fiber laser welded press-hardened steels

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Abstract
Fusion zone microstructures of fiber laser welded Al-Si coated press-hardened steels at pre- and post-press-hardened conditions were analyzed using transmission electron microscopy (TEM), TEM and nanoindentation studies suggest that dislocation-free δ-ferrite phase formed in the weld metal (by ingress of the Al-Si surface coating into the fusion zone) exhibits lower nanohardness compared to α-ferrite transformed after press hardening. The nanoscale properties of δ-ferrite, α-ferrite, and martensite and their crystallographic orientation relationships are reported.

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Boron containing press-hardened steels (PHS) are widely used for automotive crashworthy structural components such as A-pillars, B-pillars, and door rings [1–4]. Boron is added to the steel to increase its hardenability and retard the heterogeneous nucleation of ferrite at austenite grain boundaries [5,6]. After press hardening, this grade of steel reaches an ultimate tensile strength between 1500 and 2000 MPa. In order to prevent corrosion and high temperature oxidation, PHSs are often coated with various alloy coatings including Al-Si, Zn, and Zn-Ni [7–9]. Laser beam welding is usually employed to join various grades of PHS with different thicknesses and dimensions. A study on laser welding of PHS [10–14] confirmed the formation of secondary phase in the fusion zone (FZ) due to mixing of Al-Si coating in the weld pool. The secondary phase found in the FZ of laser beam welded PHS is reported to be either brittle Fe-Al intermetallic compound [10–12] or soft ferrite phase [13–15]. However, no supporting evidence for either case has been provided in the literature. These phases were demonstrated to be rich in ferrite stabilizing elements such as Al [10–15]. The existence of the brittle intermetallic compound or soft ferrite phase within the FZ of PHS was reported to be deleterious to the mechanical performance of the weld [10–15].

Despite the dramatic impact on mechanical performance, little is known about the formation mechanism of the secondary phase and its crystallographic relationship with the martensite during laser beam welding of Al-Si coated PHS. In our recent investigation [15], the formation mechanism of the secondary phases and their structure-properties relationship was systematically reported. The aim of the present study is to investigate the crystallographic and nanoscale properties of the phases formed in the FZ of laser welded PHS samples before and after press hardening.

The study was conducted on steel (thickness: 1.00 mm) with a coating composed of Al (90 wt.%) and Si (10 wt.%), having a yield strength and ultimate tensile strength of 424 MPa and 570 MPa, respectively. The composition of the used steel is reported in Table 1. The edges of 200 mm × 100 mm (length × width) sheets were milled and welded in a butt joint configuration using a IPG photons ytterbium fiber laser system (model: YLS-6000-S2) with a power and speed of 4 kW and 4 m/min, respectively; detailed description of the laser welding system can be found in previous studies [16]. The welded coupons were austenitized in a furnace at 930 °C for 5 mins and then press-hardened (at an approximate average cooling rate of 30 °C/s) between two flat dies, where a fully martensite microstructure was produced [1,17,18] with an ultimate tensile strength of above 1500 MPa. A scanning electron microscopy (SEM), transmission electron microscopy (TEM), and high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) imaging were used to study resultant microstructure. For TEM sample preparation, 3 mm discs were punched from the foil (thickness: ∼50 μm); with the FZ at the foil center; with the foils lightly etched with 2% Nital solution to observe the FZ clearly prior to punch discs. Finally, the samples were thinned by electrolytic jet polishing at 15 °C using a solution of acetic acid and 5% perchloric acid at 70 V with 60 mA. TEM study was performed in a JEOL 2010F, with an operating voltage of 200 keV. Nanoindentation was conducted using a Hysitron Triboindenter TI-900 equipped with a scanning probe.
microscope in a load control condition with a constant loading rate of 500 μN s⁻¹ up to a maximum load of 5000 μN; 10–12 indents were made on each distinct phase in the FZ and the average values are reported (the tolerance limit represents the standard deviation).

Representative SEM micrographs of as-received base metal (BM) in the pre- and post-press-hardened conditions are shown in Figs. 1a and 1b, respectively. The as-received BM microstructure delineated by two distinct morphologies of ferritic (α)-pearlitic (P) structure (Fig. 1a); the pearlite phase exists as a banded structure along the rolling direction in a ferritic matrix. The sheets were welded using the welding parameters as described in the experimental section; the FZ microstructure is presented in Fig. 1c which is referred to as a pre-press-hardened condition. The welded structure was austenitized in a furnace, followed by rapid quenching between two flat dies; the BM and FZ microstructure are shown in Fig. 1b and 1d, respectively. Typical lath martensite with evenly distributed intralath carbides (which are the product of autotempering [19]) are found in the BM after press hardening (Fig. 1b). In the pre-press-hardened condition, the FZ microstructure comprising of lath martensite (α₁′) and non-equilibrium high temperature ferrite phase (known as δ-ferrite or δ [20,21]) is observed due to transformation stresses associated with the phase transformation stresses associated with the phase transformation kinetics [23] by decreasing the free energy change associated with the austenite to martensite transformation. The α₁′ phase embedded in a massive δ matrix with no visible traces of grain boundaries; the area fraction of each phases was estimated to be about α₁′ (40%) and δ (60%), respectively. The absence of grain boundaries also implies that δ phase formed as a primary phase at high temperature during solidification, afterward α₁′ phase appeared when temperature reaches the martensite start temperature. Some sharp δ-α₁′ interfaces with a cusp shape (shown with black arrows in Fig. 1c) can be found. It is unlikely that a mixture of slender finger-like martensite (α₂′) nucleated at prior-austenite grain boundaries, and triple point junctions after press hardening; the α grains are divided with grain boundaries (marked with red arrows in Fig. 1d) suggesting that the α phase transformation occurred mainly by the process of nucleation and grain growth. However, the martensite phase fraction was estimated to decrease from 40% to about 20% after press hardening. The α₂′ appears in this case is free of autotempered carbides which can be attributed to lower martensite start temperature. It can be noted that as quenching was done from the temperature (930 °C) which is between Ac₁ and Ac₃ where only α-ferrite and austenite are present, which allows non-equilibrium δ phase to be equilibrium during soaking for 5 mins; therefore, the remaining δ will transform to more stable α phase (i.e., the mixture of δ and α₁′ transformed to α and α₂′). As a result, a uniform distribution of α and α₂′ structure is obtained in the entire FZ of the press-hardened sample.

Crystallographic analysis of the phases present at FZ of the pre- and post-press-hardened conditions were performed using TEM with SAD patterns analysis as shown in Figs. 2a-2e and Figs. 2f-2i, respectively. It can be noted that the crystal structure of α, and δ is considered to be a body centered cubic (BCC) structure whereas α₁′ and α₂′ is body centered tetragonal (BCT) structure. During solidification, when δ is formed at high temperature dislocation networks may generate in the δ phase due to transformation stresses associated with the phase transformation kinetics [23] by decreasing the free energy change associated with the austenite to martensite transformation.

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Fig. 2. Bright-field TEM study of the FZ of pre-press-hardened sample showing: (a) a mixture of δ and α′ phase, (b) lower bainite microconstituents. Diffraction patterns of (c) δ in [111] projection, (d) α′ in [001] projection, and (e) an orientation relationship between δ and α′: [111]_δ||[001]_α′, and post-press-hardened sample showing: (f) a mixture of α and α′ phase, (g) magnified view of α-α′_2 interface. Diffraction patterns of (h) α in [012]_α projection, and (i) α′_2 in [111]_α′_2 projection.
transformation. However, at elevated temperature, these dislocations can be reorganized into sub-grain boundaries through recovery and polygonization [24]. Therefore, these sub-grain boundaries can act as sites for austenite nucleation [25] and subsequent cooling to room temperature promotes the austenite to martensite transformation; therefore, it is likely to be dislocation-free [26] as observed in Fig. 2a. Sample tilting was conducted in order to confirm that only a low density of dislocations was present in this phase. This specific feature differentiates it from the transformed α phase after press hardening as observed in Fig. 2g, where a high density of dislocations can be observed on the α phase and in the vicinity of the α-α′ interfaces. Bright-field images of δ and α′ and their corresponding SAD patterns taken from the regions (c), (d), and (e) of Fig. 2a are shown. The morphology of δ observed in Fig. 2a is consistent with the SEM micrograph presented in Fig. 1c. The α′ structure found in the FZ consists of two different kinds of laths which can be classified as coarse (width 900–1100 nm) and fine (100–300 nm) laths, depending on their thickness [27,28]. A high density of dislocation in the order of 10^15 m^-2 [19] is observed which agrees well with the literature in the case of low carbon lath martensite [29]. SAD patterns that were obtained from δ and α′ were indexed based on a BCC crystal structure with a derived zone axis of [111]_α′ and [001]_α′, respectively. The lattice parameter of δ was calculated from the TEM diffraction spots to be (a) 2.934 ± 0.011 Å and martensite α = 2.925 ± 0.018 Å and c = 2.925 ± 0.018 Å; it can be noted that the calculated α- and c-axis lattice parameter for martensite was found to be exactly the same representing a negligible tetragonality (i.e. low c/a ratio). The diffraction spots corresponding to retained austenite that may exist at the interlath locations [30,31] were not detected, which implies that the cooling rate [16,32,33] was sufficiently fast to prevent carbon partitioning into retained austenite phase during laser beam welding. In order to determine the orientation relationship (OR) between δ and α′, SAD pattern was taken from the 6-α′ interface (zone (e) in Fig. 2a) confirmed the OR[111]_δ//[001]_α′. The δ phase had an angular deviation of about 14° between (111) and (001)_α′ projection planes. In addition to δ and α′ phases, a small fraction of lower bainite like microconstituents also revealed via TEM observation (Fig. 2b). The carbides with a few nanometers size were detected in the bainitic ferrite sheaf (sheaf width: 0.5–2 μm). However, detailed characterization of the lower bainite was not performed owing to their low fraction.

The post-press-hardened welded coupon was characterized using TEM and corresponding SAD patterns were obtained from each phases as represented in Figs. 2f–2l. A slender finger-like α′ phase which is found to be highly dislocated is embedded in a α matrix. The interfaces between α and α′ phase is outlined with dotted lines in Fig. 2g; a pile of dislocations is observed at the α-α′ interface as indicated by the arrows. The martensite phase observed in this kind of sample is not typical lath martensite; the common features of martensite such as laths, blocks, and packets [34] are rarely visible in this case. SAD patterns that taken from both the α and α′ phase were indexed as a BCC and BCT structure showing the projection planes of [012]_α and [111]_α′, respectively.

The FZ microstructures were further characterized using HAADF-STEM analysis as presented in Fig. 3. HAADF-STEM images were obtained by correcting z-contrast together with diffraction contrast which clearly revealed δ, lath kind of α′, α, and finger-like α′′ phase. In addition, other microstructural features such as micro twins (T) in martensite and a pile of dislocations in the vicinity of interfaces are also visible. The δ phase shown in Fig. 3a is free of dislocations due to high transformation temperature [26]. On the other hand, a dislocated α phase is found in the post-press-hardened sample (Fig. 3b); Fig. 3c shows the magnified view of α-α′ interface where an area with highly dislocated structure is highlighted with arrows. The dislocations introduced into α phase and α-α′ interfaces due to the effect of a large residual stress generated by means of the volume change during austenite to martensite transformation [35]. The compositional analysis carried out in HAADF-STEM mode shows that the δ phase is rich in Al (2.52 ± 0.08 wt%) which is decreased to 1.70 ± 0.21 wt% in α phase after press hardening; while the Si content (about 0.71 ± 0.07 wt%) remains similar for both of the conditions. From the elemental analysis, it may be reported that the diffusion of Al occurred during press hardening; Kang et al. [36] also demonstrated a similar phenomenon. The diffusion of Al increases the carbon activity and its diffusivity in austenite; therefore, a uniform distribution of α′ phase is observed in a α matrix after press hardening (Fig. 1d, and Figs. 2f and 2g). Unlike, α′ phase that has typical lath kind of morphology, the α′′ phase contains a few micro twin-like features (highlighted with yellow arrows in Fig. 3b) which are formed in order to accommodate the distortion energy during martensitic transformation [19].

In order to assess the nanomechanical properties of the phases that presents in the FZ of pre- and post-press-hardened samples, instrument nanoindentation study were conducted. The load-depth (P-h) curves and their corresponding indent impressions are shown in Fig. 4. The nanohardness measured on α′ phase possesses highest value ranges from 7.14 to 9.06 GPa, with an average of 7.88 ± 0.82 GPa. Conversely, both the ferrite phases were identified to have similar nanohardness (δ: 4.96 ± 0.21 GPa, and α: 5.05 ± 0.42 GPa); however, δ phase deformed more severely (average indent depth: 150.14 ± 5.56 nm) compared to α phase (average indent depth: 142.57 ± 5.19 nm) due to lack of dislocations as observed via TEM and HAADF-STEM study (see Fig. 2a and 3a). The initial part of the indentation curves is plotted inset of Fig. 4a; the indent impression on the δ phase shows pop-in at 9.14 ± 3 nm (indent load: 135 ± 50 μN) whereas this value extended to 22.72 ± 10 nm (indent load: 467 ± 35 μN) for the α phase. The pop-in behavior was confirmed by plotting a Hertzian elastic contact solution according to references.
Nanoscale hardness of the phases present in the FZs of laser beam welded PHS steels showing: (a) load-depth (P-h) curves for the phases of $\alpha_1$, $\alpha$, and $\alpha_2$, (b) and (c) their corresponding indent impressions.

References

[28, 37, 38] in Fig. 4a. The P-h curve on $\delta$ phase shows a deviation from the Hertzian elastic solution at very early stage of loading representing fully plastic deformation. On the other hand, the indent impression on $\alpha_2$ phase shows elastic deformation up to 22 nm depth and 467 $\mu$N load. The differences in pop-in behaviors is likely to be the influences of dislocation density which may impede the applied load. Conversely, the P-h curves of $\alpha_1'$ and $\alpha_2'$ shows similar trends up to about 3000 $\mu$N load afterward the indent on $\alpha_2'$ phase shows a pop-in at a depth of 85 ± 12 nm due to being closed to the $\alpha$-$\alpha_2'$ interface (Fig. 4c). Unlike, the indent on $\alpha_1'$ does not experience a pop-in as it located on a fine lath and shows an average nanohardness value of about 7.88 ± 0.82 GPa (Fig. 4b), which is higher than the one obtained on the $\alpha_2'$ phase (7.14 ± 0.61 GPa). The nanohardness value of the martensite phase decreased after press hardening due to diffusion of martensite hardenables elements such as C and Mn which are estimated to be lower at $\alpha_2'$ phase compared to $\alpha_1'$ ones.

In conclusion, the fusion zone microstructure of fiber laser welded press-hardened steels were characterized using TEM, HAADF-STEM, and nanoindentation study at pre-press-hardened and post-press-hardened condition. TEM analysis reveals a predominant non-equilibrium high temperature $\delta$-ferrite, martensite, a very negligible amount of lower bainite structure at pre-press-hardened condition, and a mixture of $\alpha$-ferrite and martensite structure at post-press-hardened condition. The $\delta$-ferrite phase is found to be dislocation-free whereas $\alpha$-ferrite, and $\alpha$-ferrite - martensite interface contains a pile of dislocations. The nanoscale hardness testing revealed early pop-in behavior on $\delta$-ferrite phase suggesting it is slightly softer than the $\alpha$-ferrite due to the absence of dislocations.

Fig. 4. Nanoscale hardness of the phases present in the FZs of laser beam welded PHS steels showing: (a) load-depth (P-h) curves for the phases of $\alpha_1'$, $\alpha$, and $\alpha_2'$, (b) and (c) their corresponding indent impressions.