

Research paper

Microstructure, mechanical properties and hydrogen embrittlement behaviour of wire arc additive manufactured AISI 316L under high-pressure gaseous hydrogen environment

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ABSTRACT

Hydrogen is highly alluring globally due to its role in the renewable energy revolution and decarbonization potential. However, effective hydrogen storage poses substantial challenges in materials, as they are inclined to failure. In general, AISI 316L is preferred for hydrogen storage due to its resistance to Hydrogen Embrittlement (HE) behavior. To effectively control HE-based failures, a new material category developed via Wire Arc Additive Manufacturing (WAAM) using Cold Metal Transfer (CMT) technology is proposed. The significance of CMT technology lies in its low heat-generating ability and high arc stability. As a result, the likelihood of austenite-to-martensite transformation is reduced, allowing the material to retain the FCC phase. CMT process in terms of its low heat input characteristics. Which prevents the material from becoming brittle and reduces the risk of failure in a hydrogen environment. This work develops additively manufactured AISI 316L samples with linear and weaving deposition patterns to investigate failure behaviour. The samples are characterized before and after gaseous hydrogen charging at 80 bar using a Sieverts apparatus. The optical imaging results indicate that the weaving pattern of metal-deposited samples maintains higher austenite phase stability than the linear pattern. The UTS of the hydrogen-exposed linear- and weaving-pattern samples are 765 MPa and 801 MPa, respectively. EBSD results revealed that the hydrogen-exposed weaving pattern does not affect grain boundaries or the grain internal state. Based on various analyses in a hydrogen environment, weaving patterns are found to be highly resistant to HE and maintain the ductility of the as-printed samples.

1. Introduction

Addressing the energy crises is crucial to supporting sustainable development. In this regard, the entire transportation sector may need to completely transition away from non-renewable energy, particularly fossil fuels, toward renewable energy sources in the near future. Among them, hydrogen stands out as an inevitable choice due to its unique features, including decarbonization, cost-effectiveness, environmental cleanliness, and the ability to be produced on demand [1]. This positive impact stems from hydrogen itself; however, poor infrastructure development poses a significant challenge to its effective utilisation. Therefore, developing an effective infrastructure for hydrogen storage is

essential. Nowadays, research efforts focus on improving hydrogen storage capacity through advancements in modern materials and fabrication methods like additive manufacturing (AM) [2]. Despite the numerous benefits of hydrogen, limitations in efficient storage, transportation, and distribution pose significant materials-related challenges, as these systems are prone to failure [3]. These limitations include hydrogen's highly flammable nature in the atmosphere, necessitating careful handling [4]. In metallic materials, hydrogen-induced degradation occurs when hydrogen atoms diffuse into the atomic lattice, weakening atomic interactions and leading to progressive losses in ductility and fracture toughness. This phenomenon is known as HE [5]. For hydrogen storage materials, HE is a well-documented cause of

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hydrogen-induced failure, particularly concerning Stainless Steel (SS) in industries, automotive, aerospace, and other critical applications [6]. Hydrogen atoms diffuse into the material surface, infiltrating the crystal lattices, thereby altering the crystal structure and affecting mechanical and tribological properties, which depend on the materials and their alloying elements, as well as environmental conditions [7]. Compared to other materials or alloys, AISI316L and AISI304 are preferred for hydrogen storage devices due to their higher HE-resistant behaviour and Ni-to-Cr equivalent ratios, which help maintain phase stability in a hydrogen environment. In particular, AISI 316L materials exhibit superior resistance to HE compared to SS304, owing to the effects of molybdenum (Mo) addition [8]. The incorporation of Mo addition into iron (Fe) and nickel (Ni) greatly improves corrosion resistance in both gaseous and electrochemical environments. AISI 316L contains 2.42 wt % Mo, while AISI 304 has only 0.360 wt%. Additionally, the presence of Ni is also significantly higher in SS316L compared to SS304, which contributes to stabilising the austenite phase (FCC) structure [9].

Moreover, the addition of non-metallic elements, such as carbon, also influences HE behaviour. Generally, a lower carbon content in the alloying system leads to the formation of precipitates and carbide phases during heat treatment or welding, thereby enhancing corrosion resistance in steel-based materials [10]. Conversely, some research suggests that a higher carbon content in the materials can help resist HE and control hydrogen-induced fracture behaviour [11]. This improvement is ascribed to the cohesive effects of carbon enrichment at grain boundaries, which enhance cohesion and help prevent HE failures in austenitic steel [12]. These important properties are further enhanced through modern material fabrication methods, like Metal Additive Manufacturing (AM) [13]. In recent days, high research attention has focused on the AM: WAAM process. Compared to other AM processes, CMT has high intensity because it is coupled with a reduced heat input to maintain better arc stability and an innovative wire feeding system, which improves the overall printing efficiency compared to other AM techniques [14]. Supplying a high heat input can lead to a transformation from a metallic to a brittle intermetallic phase; however, these negative effects are minimised by utilising the CMT method. Interestingly, a lower heat input correlates with increased microhardness and improved interfacial characteristics. The higher heat input to the materials increases the likelihood of developing an intermetallic phase fraction [15].

The Sieverts apparatus is a commonly used and trustworthy method for assessing a material's ability to absorb hydrogen. It is one of the most widely used tools for accurate hydrogen storage characterization because it provides excellent measurement precision with a straightforward, affordable, and durable design [16]. Based on hydrogen absorption/desorption evaluation, only limited studies are available, such as calorimetric, gravimetric, volumetric, and temperature-programmed desorption (TPD) [17]. The calorimetric methods are based on supplying hydrogen to a chamber, where the samples absorb hydrogen atoms, followed by an exothermic reaction (heat release), causing the samples to release hydrogen [18]. The gravimetric method measures the hydrogen absorption/desorption rate based on weight, or the microbalance method. Based on this density uncertainty, the author noted that samples change dimensions (expand) in the hydrogen environment; based on these dimensional changes, the hydrogen-absorbed samples are expected to change the dimensions of the materials. These results were also confirmed by simulation techniques to be $\pm 25\%$ deviations in hydrogen-absorbed/desorbed samples [19]. The volumetric method for determining the hydrogen absorption/desorption behaviour of the materials is based on manometer measurements, similar to the change in gas volume influenced by hydrogen absorption in the samples [20]. However, TPD is used to measure the hydrogen desorption rate relative to the amount of absorbed hydrogen at a constant temperature. Due to the use of lower density materials, such as the addition of graphene nanoparticles (GNPs), single- and multi-walled carbon nanotubes (CNTs), lithium, carbide, and

nitride, the Sieverts apparatus is limited in its ability to assess hydrogen absorption rate [21].

In hydrogen storage and transportation, hydrogen-induced material failures occur due to ineffective fabrication techniques, such as high heat input, poor cooling, rapid thermal cycling, and high residual compressive stress, which lead to the development of an alpha martensite phase. This martensite phase is highly hard and is prone to crack initiation in the hydrogen storage environment. So, to control the martensite phase formation in AM, with the help of a newly developed fabrication method called CMT [22]. Compared to conventional MIG welding, the CMT process utilises only 10 % of the heat to print the materials, so the process is called a cold process [23]. The primary objective of employing the WAAM process is to achieve highly efficient metal deposition using conventional materials and equipment, thanks to its short-circuit effect. The CMT technology involves considerably reduced raw material usage (50 to 80% lower) for deposition, as well as significantly lower heat application (reducing heat input by 5 to 10 times) while developing oxide-free components, less distortion, zero spatters and the wire deposition rate 2-3 kg/h [24]. During material fabrication, controlling heat input is crucial for achieving optimal phase structure and microstructural properties in CMT technology, with heat input ranging from 60 to 120 J/mm [25].

Materials resistant to HE are particularly preferred when they exhibit lower hardness levels [26]. Given these factors, AISI 316L is a predominant material choice for high-pressure hydrogen gas storage, applicable in type 1 (fully metallic materials) or type 3 (metallic liners) pressure vessels. During high-pressure hydrogen storage, these linear materials sustain approximately 20% of the load while also managing gaseous hydrogen permeability to mitigate hydrogen-induced failures [27]. To address these hydrogen-induced failures effectively, advancements in materials development, design strategies, and fabrication methods are needed.

Among various material manufacturing methods, melting base powders or solid materials helps achieve the desired alloy compositions. They have to undergo melting, solidification, cooling, reheating (stress-relieving), and subsequent cooling, which involves a solid-state phase transformation. The cyclic heating and cooling process causes the material to expand and contract, generating internal residual stress. The applied temperature is the inhibiting factor for the development of this residual stress. During the WAAM process, high residual compression stress can lead to internal cracking, brittle fracture, and material warping. In this process, residual stress is categorised into 3 types: type 1 (macroscopic deformation), type 2 (grain/texture deformation), and type 3 (atomic-level or dislocation). Compared with types 2 and 3, type 1 (macroscopic deformation) appears to be predominant in WAAM [28].

In this novel work, the aim was to evaluate the performance of wire arc-additively manufactured AISI316L with linear and weaving patterns in terms of HE behaviour under 80 bar pressure and 480 hours of gaseous hydrogen exposure in the Sieverts apparatus, using the gravimetric method (volume change behaviour) at room temperature. X-ray diffraction analysis identified the single-phase major austenite and a limited ferrite phase on the exposed material. The austenite and ferrite phase distributions are visually identified with the help of optical microstructure analysis. The surface features of the WAAM material are characterised using a high-resolution field-emission scanning electron microscope. The elemental distribution analysis identified the various elements found in the as-built samples. Electron backscattered diffraction analysis was performed on the as-printed samples to determine the grain types (linear and weaving patterns), grain boundaries, misorientation angle distributions, and their grain internal states. The effects of hydrogen atom distribution and their elastic and plastic behaviour are identified through a stress-strain diagram obtained from a tensile test.

2. Experimental methodology

In this work, SS316L filler wire with a wire diameter of 1.2 mm and

substrate thickness of 7 mm was deposited over the SS304 plate. Although SS304 has a low carbon content compared to SS 304L, the carbon dilution from the substrate to the deposited SS316L is negligible due to the low heat input and limited dilution associated with the CMT process. However, the same austenitic phase is used for both the substrate and the deposited material to avoid thermal expansion, which can lead to cracks and delamination between the built and deposited materials. The metal is deposited using a pure argon shield (18 L/min) comprising 98% argon and 2% CO₂. A 45 mm bidirectional path is utilised, with the welding current set at 70 Amps, voltage at 15.4 V, and speed at 55 cm/min. The wire feed rate was 1.9 m/min, yielding a deposition rate of 2.2 kg/h for SS 316L. The current, voltage, wire-feeding speed, substrate, and interlayer temperatures are important parameters for successful component development. The height of the as-printed parts is related to the input current during material fabrication. The CMT-based low-current supply produced stable, fine, straight-build parts with reduced material overflow. A 6-axis Fronius Cold Metal Transfer (CMT) robot prints the materials while maintaining a 16 mm tip-to-workpiece distance. Based on the heat input, current, voltage and torch travel speed, an arc efficiency of 85% was achieved. Substrate preheating is required to develop a smooth surface, increase adhesion bonding strength, and control crack and distortion effects. This results in a lower thermal gradient, which controls the formation of internal residual stress. To minimise the degree of sensitisation (DOS) of the deposited material, the interpass temperature is maintained at 150°C. The interpass temperature controls heat accumulation and reduces excessive heat build-up, thereby increasing tensile stress and improving the ductility of as-printed parts. During the layer-by-layer metal deposition, once a layer is deposited, the surface is cleaned using a surface cleaning agent (acetone), to remove the unwanted surface impurities present. Subsequently, the as-printed parts are subjected to a surface treatment process using Kalling's reagent, comprising non-ionised water (40 ml, copper chloride (CuCl₂) 2 grams, hydrochloric acid 40 ml, and combinations of ethanol (85%) + methanol (15%) (40 -80 ml. Fig. 1 shows the WAAM-printed (i) linear and (ii) weaving pattern samples. The materials are then cut using a Job Master D-Zire wire-cut electrical discharge machine, achieving a high surface finish of 0.6 µm.

In recent days, there are various experimental methods available for hydrogen-related research conducted on advanced materials, such as hydrogen charging methods, such as high-pressure gas hydrogen charging [29], autoclave hydrogen charging [30], thermal gas charging [31], high-temperature, high-pressure hydrogen charging [32], Sieverts apparatus-based hydrogen charging [16], and In-situ mechanical hydrogen gas charging [33]. Based on various studies using different methods/devices, the Sieverts apparatus appears to be an effective method for evaluating material properties and performance. The Sieverts apparatus determines the hydrogen absorption capacity of a

material using a volumetric calibration method based on pressure measurements. Initially, hydrogen gas is introduced into the reference chamber under controlled pressure and room-temperature conditions, with a hydrogen purity of 99.999%. The gas is then allowed to expand into the sample chamber containing the test specimen. As the specimen absorbs hydrogen, the pressure within the system decreases. The hydrogen uptake is subsequently calculated from the pressure difference between the reference and sample chambers using the calibrated chamber volumes and the gas laws. This approach enables accurate and reproducible determination of the hydrogen absorption capacity of the material. The hydrogen charging test was conducted at an initial pressure of 80 bar for 480 h. At the end of the experiment, the pressure was maintained at 78.80 bar, corresponding to a pressure loss of only 1.20 bar. This minimal pressure reduction indicates good pressure stability throughout the test under room-temperature conditions (26.5°C). Fig. 2 presents the methodology adopted for hydrogen performance evaluations in materials under the Sievert's apparatus. The material property evaluation is based on ex-situ tensile test measurements to determine the stress-strain behaviour of as-printed samples. Further, the material performance has been evaluated based on hydrogen absorption/desorption to determine storage capacity, and pressure-composition-temperature (PCT) conditions have been evaluated. In particular, the scope of this work is to investigate the material properties of WAAM-SS316 under gaseous hydrogen charging. In addition, the performance of the as-printed materials with different weaving patterns in terms of hydrogen absorption/desorption rate, storage capacity, Hydrogen-to-Metal ratio (H/M), and pressure-temperature conditions is evaluated. Fig. 3 shows the experimental setup of the Sievert's apparatus used for this research work.

X-ray diffraction (XRD) analysis was used to evaluate the deposited materials for phase information at a scanning rate of 4° min⁻¹. Additionally, microstructural and elemental quantification, along with crystallographic texture information regarding the samples, were obtained using a Scanning Electron Microscope (SEM) equipped with Energy-Dispersive Spectroscopy (EDS) and Electron Backscatter Diffraction (EBSD). The samples were prepared according to ASTM E3 standards. The step size and angle of inclination were 1.75µm and 70°, respectively, for EBSD analysis. The stress-strain behaviour of hydrogen-exposed and non-hydrogen-exposed samples was evaluated using a uniaxial tensile test in accordance with ASTM E8, with a tensile rate of 0.5 mm/min. The effects of hydrogen absorption and its mechanical properties on linear and weaving-type patterns were evaluated. The residual stress behaviours of the as-printed materials are assessed using a Pulstec Industrial standard µ-X360 equipment. The entire Debye-Scherrer ring is captured at a single fixed incident X-ray angle using the cos-α approach, which forms the basis of the study. The Kα X-ray tube, for example, operates at a voltage of 30kV and a current of 1mA. X-

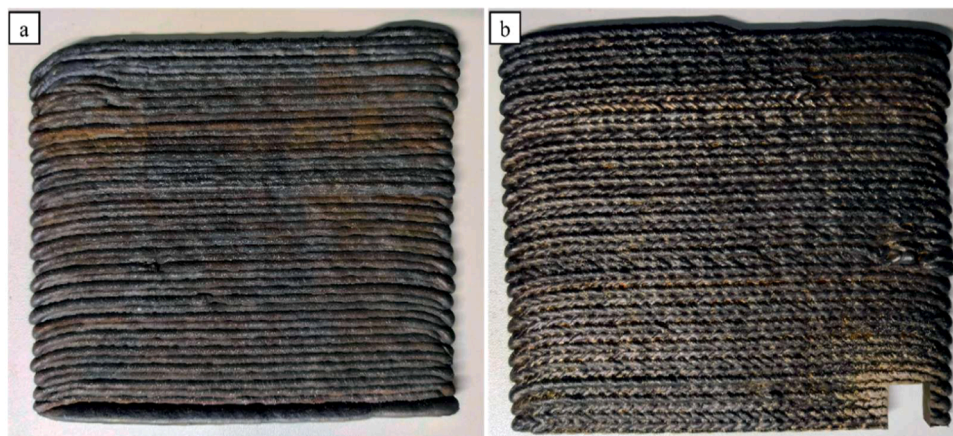


Fig. 1. As-printed WAAM manufactured AISI316L material (a) Linear pattern (b) Weaving pattern.

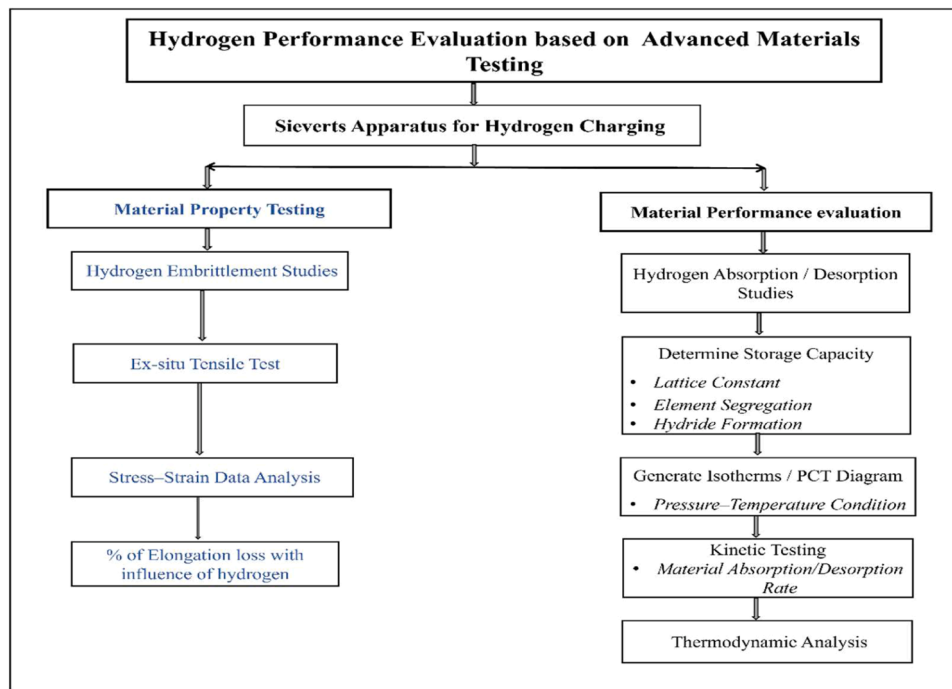


Fig. 2. Methodology adopted for hydrogen performance studies using the Sieverts apparatus.

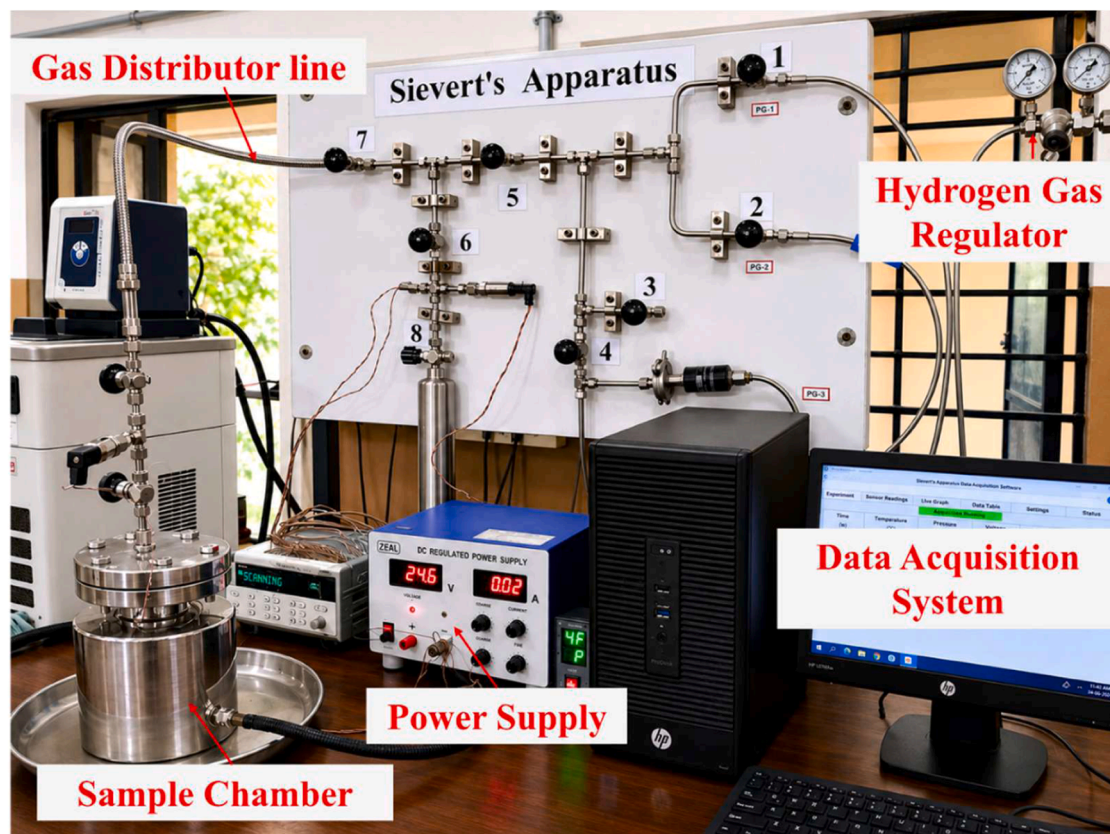


Fig. 3. Experimental setup.

rays are concentrated on 1.016 mm collimator lenses. Using the single incidence angle approach, the X-rays are directed towards the target. The measurement takes 90 seconds. The specimen-to-detector distance was 50 mm, and the beam incident on the sample was approximately

28.8°. The as-printed samples have a diameter of two millimetres and an X-ray penetration depth of five micrometres. The hardness of the samples was measured using Vickers microhardness, in accordance with ASTM E384.

3. Results and discussion

3.1. XRD analysis

In general, materials with the iron FCC (austenite) phase exhibit higher resistance to HE failures than those with the iron BCC (ferrite) phase, due to their greater ductility. Compared to conventional manufacturing, the WAAM AISI 316L material shows a higher FCC phase than the BCC phase. Because of its lower heat input to melt the materials, based on CMT technology [34]. Fig. 4 represents the XRD analysis of the linear and weaving patterns of WAAM AISI 316L samples. Here, the Bragg peaks correspond to the crystal structure. In both linear and weaving spectra, similar forms of iron are observed. Based on these peaks, the intensity indicates that the WAAM materials may exhibit a major FCC iron (austenite) phase with a minor BCC iron (ferrite) phase in both linear and weaving configurations [35,36].

3.2. Performance of linear and weaving patterns, their microstructure and EBSD analysis

The effects of heat supply are important considerations for melting the filler wire and for interface behaviour across various WAAM-based pattern developments. In this direction, the material is melted using CMT to achieve extremely low heat input and high arc stability. For better microstructure, arc stability is critical, and it depends on the CMT process, which uses reverse polarity to develop a stable arc at very low temperature. This welding process involves (i) peak and background current phases and (ii) a voltage short-circuit phase. Based on better mechanical property performance, the short-circuit phase retracts the filler, then breaks the circuit, reducing heat input at the time of joint (layer-by-layer deposition). In this type of lower-heat-input control, an increase in intermetallic layer thickness is followed by improved joint performance in the linear- and weaving-pattern-built components. With lower heat input, the FCC (austenite) phase prevents the BCC (ferrite) phase transformation, thereby mitigating the initial onset of material brittleness. Accordingly, the as-printed materials exhibit a higher dislocation density, maintain similar crystalline size development across the alloying elements, and avoid precipitation. For this reason, hydrogen-induced failures are controlled to enhance the material's ductility. Fig. 5 represents the optical microstructure analysis of AISI 316L (a) linear and (b) weaving patterns. Based on microstructural analysis, a lower amount of ferrite phase followed by a higher amount of austenite phase is visible in the weaving pattern compared to the linear pattern.

Fig. 6 shows the scanning electron microscope analysis of WAAM samples (a) linear and (b) weaving patterns. Fig. 7 presents quantitative

data on various elements, including detailed confirmation from Energy-Dispersive X-ray Spectroscopy (EDS). The properties of as-printed materials depend on factors such as element deposition, diffusion length, and Inter Metallica Width (IMW). This work focused on the as-printed material samples and their elemental diffusion rate, which are strongly influenced by the heat input. Element diffusion behaviour is represented by elemental distribution analysis. These properties are influenced by heat input, particularly through the WAAM-CMT technology. The effects of feed wire deposition with various patterns on the initial elemental composition are shown in Table 1, which presents the feed wire and the as-printed linear and weaving-pattern material compositions. The combined effects of elemental segregation during rapid solidification, dilution between successive deposited layers and the substrate, and chromium's high oxygen affinity, which encourages limited oxidation during deposition, are responsible for the decrease in Cr content from 16.0 weight per cent in the feed wire to 12.7 weight per cent and 14.6 weight per cent in the linear and weaving deposition patterns, respectively. Similarly, elemental redistribution and dilution brought on by the WAAM process's multiple heat cycles are linked to the decrease in Ni concentration from 12.8 weight per cent to 10.9 weight per cent and 11.4 weight per cent.

The primary cause of the observed variation in Mo concentration is its propensity to segregate into interdendritic areas during solidification, which results in localised compositional changes that EDS can detect. Mn's relatively high volatility under the high arc temperatures of the WAAM process, which encourages partial elemental evaporation during deposition, is mostly responsible for the drop from 2.2 weight per cent to 1.3 weight per cent and 1.9 weight per cent, respectively, for the linear and weaving patterns. It should be mentioned that these EDS data do not indicate substantial changes in the bulk alloy composition, but rather local compositional differences resulting from the temperature history of the deposition process.

Materials with a higher Ni content favour the formation of the austenite phase. Followed by the influence of the fabrication method and the carbon content of the as-printed part. In this regard, the weaving pattern used to control the development of carbon percentage is the WAAM product, as shown in the EDS analysis.

In particular, the elements Cr and Ni are highly resistant to HE. The addition of Cr, which is highly corrosion-resistant and provides strong protection against oxidation, and Ni stabilises the FCC (austenitic) phase structure. Following the addition of Ni and Cr metallic elements to Fe, the diffusion effects are significant in Fe and Ni. The higher diffusion of Fe into Ni results in higher metal deposition, followed by an increase in the Intermetallic Width (IMW). A higher IMW leads to a more stable FCC (austenite) phase, increasing the ductility and toughness of the as-printed material. High ductility and toughness help resist the materials HE. In this regard, compared to the linear pattern, the weaving pattern is found to be superior [26].

The EBSD results also provide phase information (austenite and ferrite) and surface texture information (grain and grain boundary features). The phase formation and transformation of iron-based materials depend on their melting temperatures. In this regard, WAAM-CMT technology ensures low heat input and high arc stability. The heat input effectively refines the grains and maintains a stable phase structure. Considering this, the EBSD results also show that CMT-printed materials maintain high phase stability. The EBSD phase results for the linear and weaving patterns showed 98.7% FCC (austenite) and 1.3% BCC (ferrite) in both cases. The formation of 1.13 wt.% δ -ferrite is attributed to the rapid solidification and low heat input of the WAAM-CMT process. The repeated thermal cycles and the Cr/Ni balance of AISI 316L promote the retention of a small amount of residual δ -ferrite, while the microstructure remains predominantly FCC (austenitic).

Fig. 8 shows the EBSD representation of hydrogen-exposed sample with a linear pattern: (a) band contrast (BC) + Inverse Pole Fig. (IPF) + Grain Boundary (GB) and (b) Grain Internal Statistics (GIS).

The various EBSD maps in BC, IPF, GB, and GIS represent

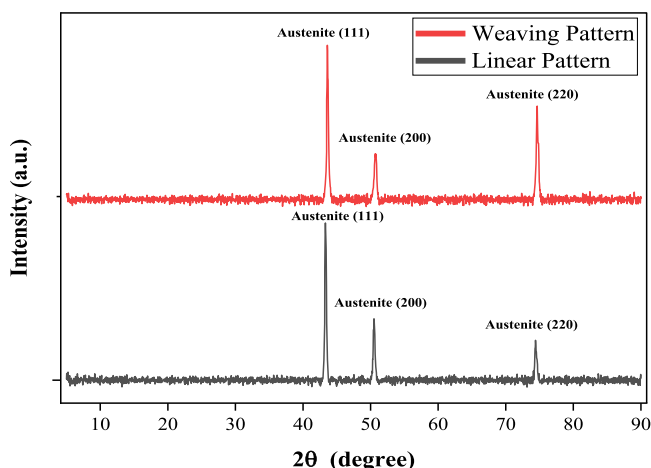


Fig. 4. XRD analysis of the as-printed linear and weaving patterns.

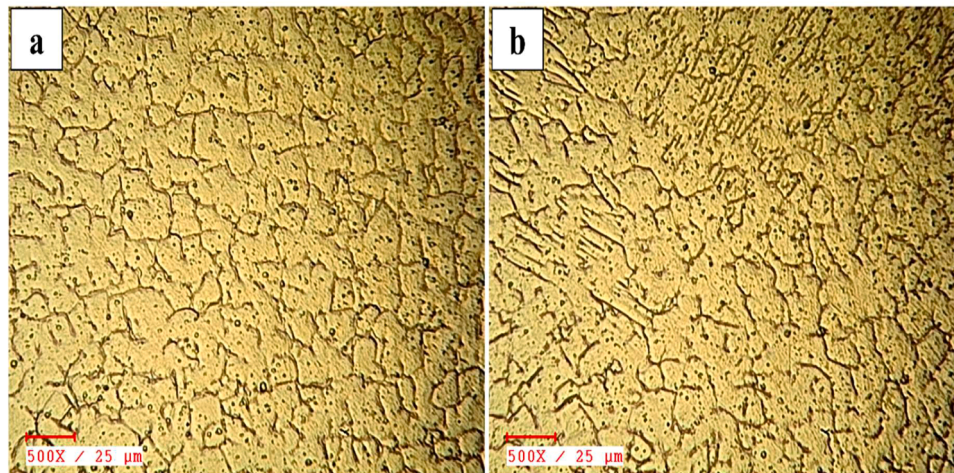


Fig. 5. Optical microstructures of WAAM-manufactured AISI 316L (a) linear pattern, (b) weaving pattern.

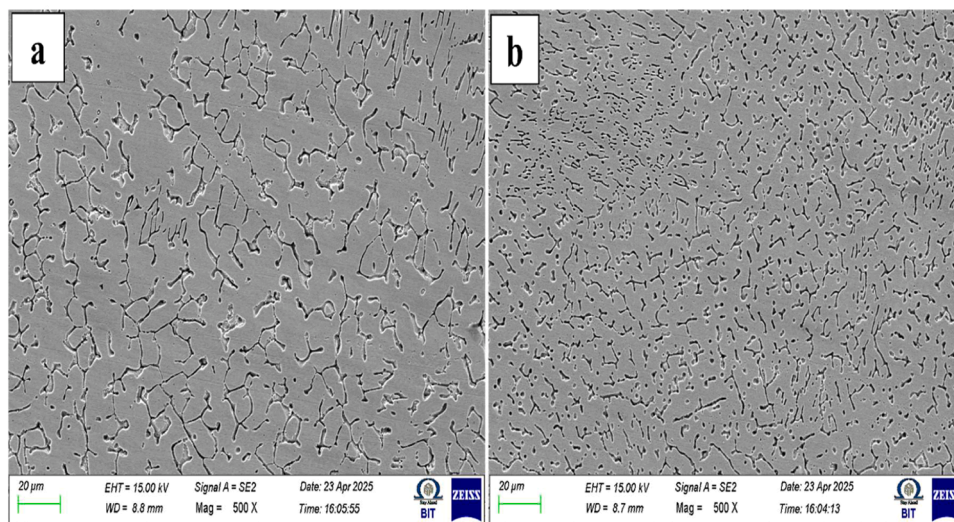


Fig. 6. Microstructure of WAAM-manufactured AISI 316L (a) linear pattern, (b) weaving pattern.

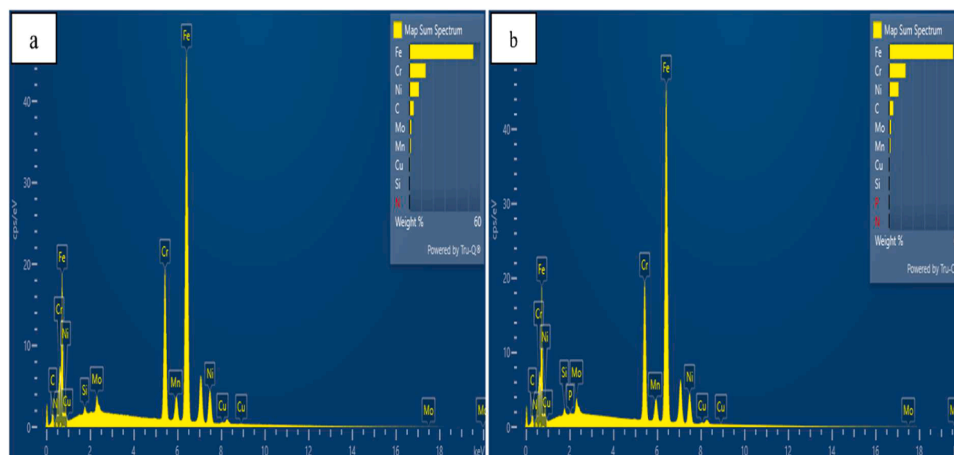


Fig. 7. EDS analysis of WAAM-manufactured AISI 316L material (a) linear pattern, (b) weaving pattern.

quantification information on boundary characteristics. Fig. 9 shows the EBSD representation of a hydrogen-exposed sample with a weaving pattern: (a) band contrast (BC) + Inverse Pole Fig. (IPF) + Grain

Boundary (GB) and (b) Grain Internal Statistics (GIS).

The various angular grain boundaries are directly related to the evaluation of recrystallised texture. In this regard, the linear and

Table 1
Elements present in WAAM-manufactured samples based on the feed wire, linear and weaving pattern.

Pattern type	Cr	Ni	Mo	Mn	Cu	Si	C	Fe
Feed wire (AISI316L)	16.0	12.8	2.1	2.2	0.1	1.1	0.9	Balance
Linear pattern	12.7	10.9	1.8	1.3	0.2	0.9	2.3	Balance
Weaving pattern	14.6	11.4	1.5	1.9	-	0.6	1.8	Balance

weaving patterns develop 59.6% and 62.7% of HAGBs, respectively. At

higher percentages, HAGBs control the as-printed materials and their dislocation motion. This lower dislocation motion enhances toughness. In this, higher toughness is used to mitigate crack propagation and increase hydrogen trapping at reversible hydrogen sites, thereby controlling HE failure. These features are represented as the grain's internal state.

The degree of intragranular deformation is indicated by Grain Orientation Spread (GOS), which is derived from EBSD analysis and used to assess the variance in crystallographic orientation inside individual grains. Fig. 10 represents the GOS analysis of (a) linear pattern

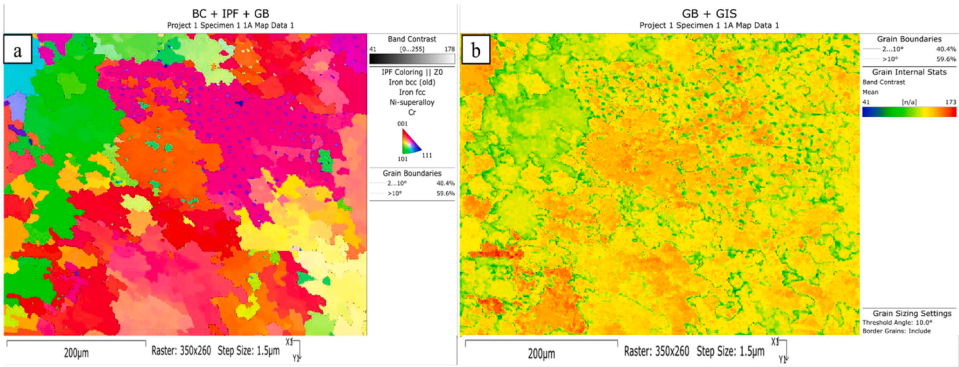


Fig. 8. EBSD analysis of a hydrogen-exposed sample with a linear pattern: (a) BC+IPF+GB features, (b) GIS features.

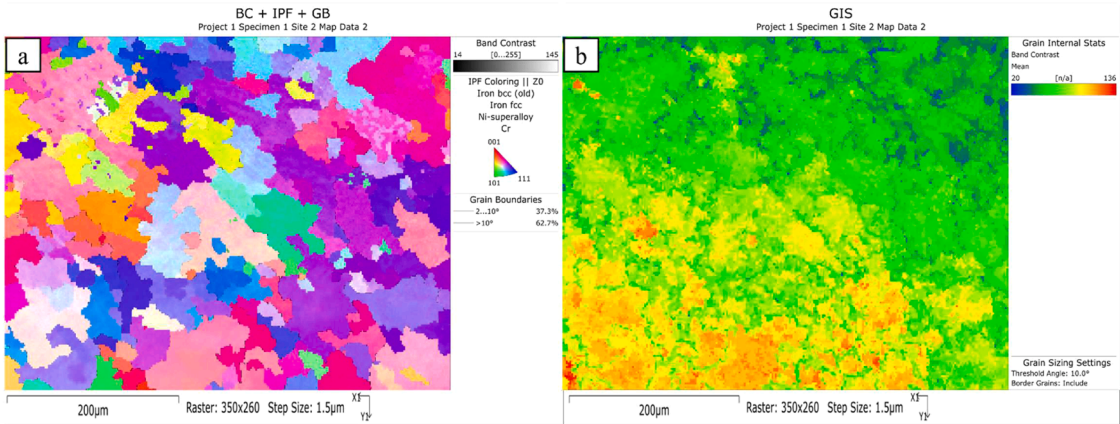


Fig. 9. EBSD analysis of a hydrogen-exposed sample with a weaving pattern (a) BC+IPF+GB features, (b) GIS features.

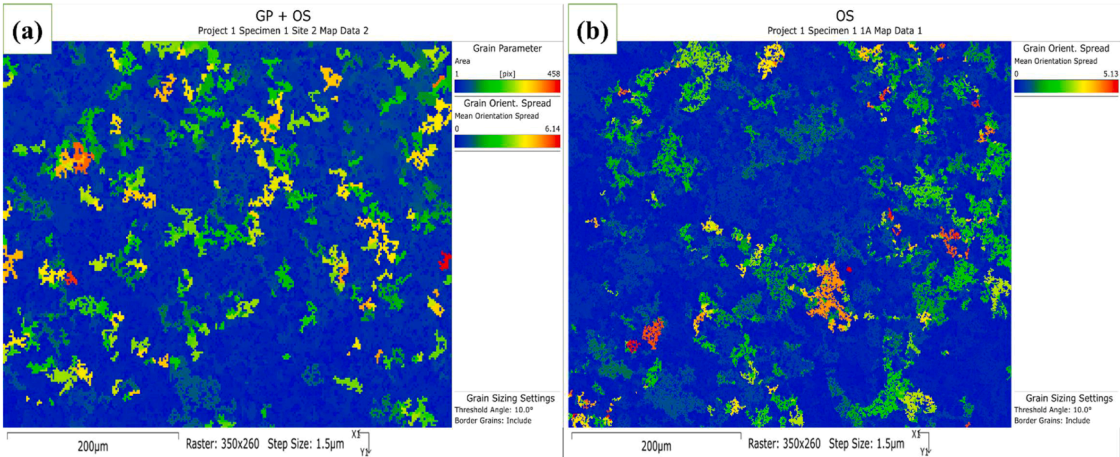


Fig. 10. Grain orientation Spread analysis of (a) linear pattern, (b) Weaving Pattern with hydrogen-exposed sample.

and (b) weaving pattern samples in a hydrogen-exposed condition. Grains with relatively homogeneous crystallographic orientations, low internal strain, and restricted plastic deformation are often indicated by GOS values $\leq 5^\circ$. GOS values $> 5^\circ$, on the other hand, indicate larger orientation deviations within grains, which are indicative of increased stored strain energy, higher dislocation accumulation, and enhanced plastic deformation. The linear pattern has a GOS value of 6.14° , and the weaving deposition pattern has a GOS value of 5.15° . Based on the results, a more uniform grain orientation with less internal strain was indicated by the weaving deposition pattern's lower GOS value in the current investigation compared to the linear deposition pattern. This implies that the weaving technique has better resistance to plastic deformation, which should increase its resistance to hydrogen embrittlement. Fig. 11 show the Kernel Average Misorientation (KAM) maps analysis of gaseous hydrogen exposed (a) linear pattern, (b) Weaving Pattern.

Compared with the linear deposition pattern (4.99°), the weaving deposition pattern exhibited a lower average KAM value (4.81°), indicating reduced dislocation formation and local lattice distortion. A more homogeneous strain distribution is indicated by the lower KAM value, which could enhance resistance to hydrogen embrittlement and plastic deformation. Fig. 12 shows the $\{100\}$, $\{110\}$, and $\{111\}$ representations of pole Figs obtained from the EBSD analysis of WAAM-fabricated AISI 316 specimens deposited using linear and weaving strategies. Both samples exhibit a distinct FCC crystallographic texture, confirming the presence of preferred grain orientations after deposition. The weaving deposition pattern produced a higher maximum pole Fig. intensity than the linear pattern indicating the development of a stronger crystallographic texture. However, the weaving specimen also exhibited lower KAM and GOS values, suggesting reduced local lattice distortion and lower intragranular strain. This combination of a well-defined crystallographic texture and lower internal deformation indicates that the weaving deposition strategy promotes a more stable microstructure, which is expected to improve resistance to hydrogen-assisted plastic deformation and embrittlement.

Fig. 13 Represents the grain disorientation or misorientation angle distribution analysis of linear and weaving patterns of WAAM samples. The misorientation angle distribution results illustrate that LABs and HABs are significant factors in the mechanisms that strengthen grain boundaries. The regions of specimens have LABs (2° - 15°) and HABs ($>15^\circ$), respectively, based on the analysis with significant peaks at 40 - 45° in the linear pattern and 5 to 15 and 40 to 45° , indicating limits in the weaving pattern. HAGBs are the major grain-boundary type that determines the strengthening behaviours of as-printed materials. The higher the fraction of HAGBs, the better the grain boundary strengthening and the less likely crack propagation. Combined with the weaving

specimen's lower KAM and GOS values, the data show that the weaving deposition method is more resistant to plastic deformation and hydrogen embrittlement than the linear deposition pattern.

Based on all the EBSD results related to HE mechanisms like Hydrogen-Enhanced Localised Plasticity (HELP) and Hydrogen-Enhanced Decohesion (HEDE). The Hydrogen-Enhanced Localised Plasticity (HELP) mechanism states that the presence of hydrogen increases dislocation mobility, which causes enhanced lattice distortion and localised plastic deformation. The degree of intragranular strain and local deformation was assessed in this study using GOS KAM analysis. In comparison to the linear deposition pattern, the weaving deposition pattern's lower KAM and GOS values show less internal strain, less dislocation buildup, and less local crystallographic misorientation. These results imply that the weaving microstructure is more resistant to hydrogen embrittlement and less susceptible to hydrogen-assisted localised deformation, which lowers the risk of crack initiation. The HEDE mechanism states that hydrogen accumulates at grain boundaries and weakens atomic bonding, leading to the development of cracks in hydrogen-exposed materials. Due to their high dislocation density, LABs function as preferred hydrogen trapping sites, while HAGBs more evenly distribute stress and prevent crack development. Consequently, it is anticipated that a microstructure with a higher percentage of HAGBs and a lower percentage of LABs will exhibit greater resistance to hydrogen-induced decohesion.

3.3. Tensile stress behaviour of WAAM samples with and without hydrogen exposure

As-printed materials and their elastic and plastic behaviour are evaluated prior to fracture using the stress-strain relationship. The tensile test determines the materials' UTS and YS in the build direction. Fig. 14 shows the tensile test sample specimens for (a1) and (b1) for without- and with-gaseous hydrogen-exposed weaving-pattern samples, followed by (a2) and (b2) for without- and with-gaseous hydrogen-exposed linear-pattern samples.

Fig. 15 shows the typical stress-strain diagram for 480 hours at 80 bar pressure, for the continuous hydrogen-exposed and non-hydrogen-exposed samples. The stress-strain diagram shows that the YS and UTS of linear pattern samples under non-hydrogen-exposed conditions are 721 MPa and 795 MPa, respectively. Furthermore, for the weaving pattern samples, the YS and UTS values are 751 MPa and 819 MPa, respectively. For the hydrogen-exposed condition, the YS and UTS of linear pattern samples are 754 MPa and 765 MPa, respectively, whereas for weaving pattern samples, they are 762 MPa and 801 MPa, respectively. A decrease in UTS value due to the influence of gaseous hydrogen atoms weakens grain boundaries and increases strain under hydrogen-

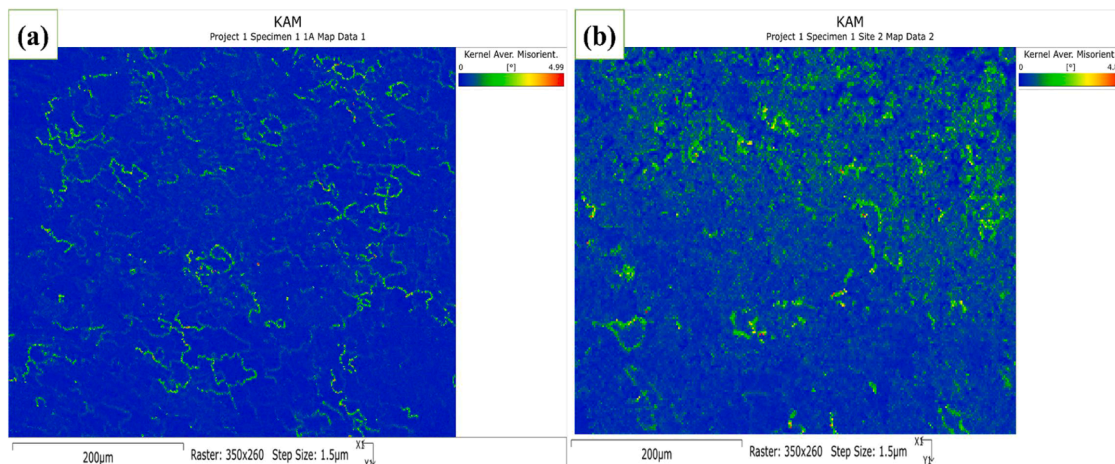


Fig. 11. Kernel Average Misorientation (KAM) maps analysis of (a) linear pattern, (b) Weaving Pattern.

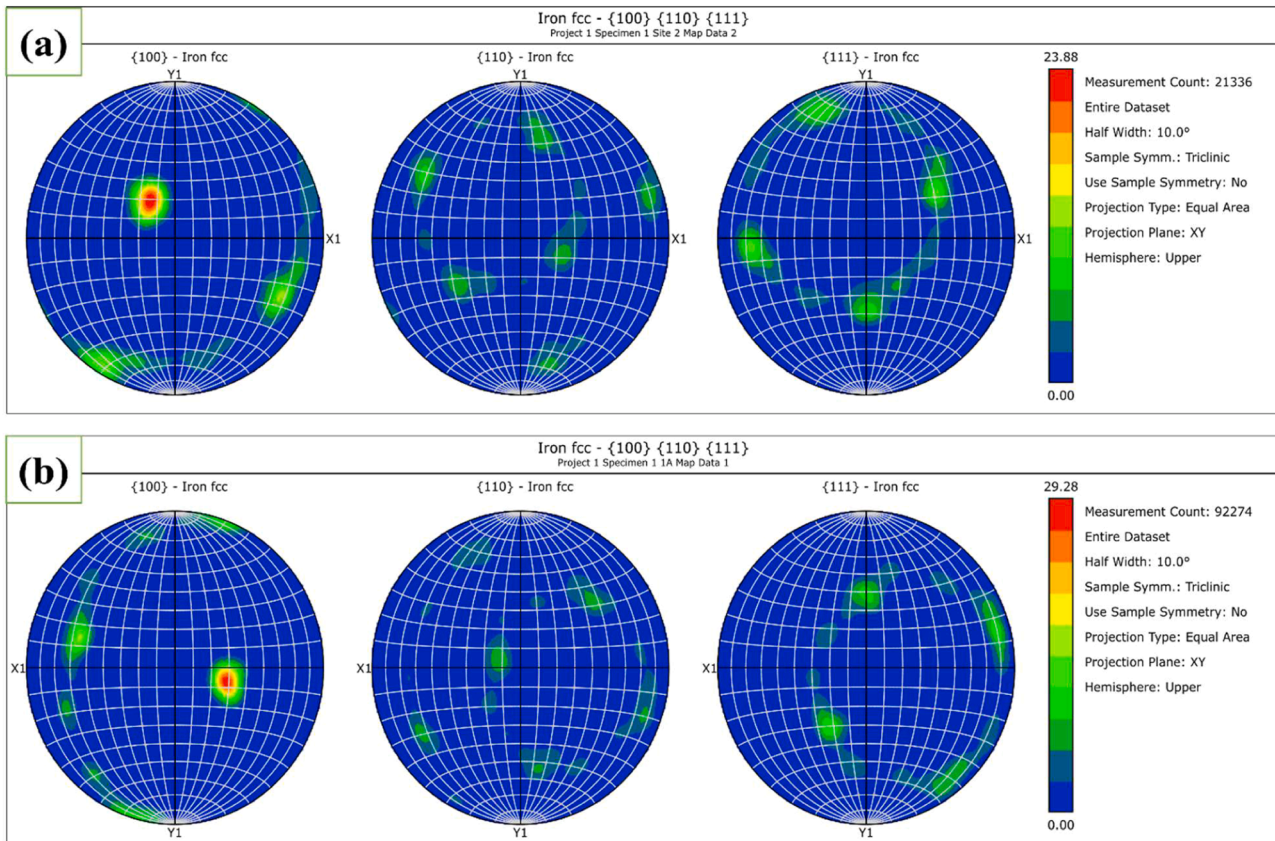


Fig. 12. Pole Fig. (PF) Distribution analysis of (a) linear pattern, (b) Weaving Pattern in a hydrogen-exposed condition.

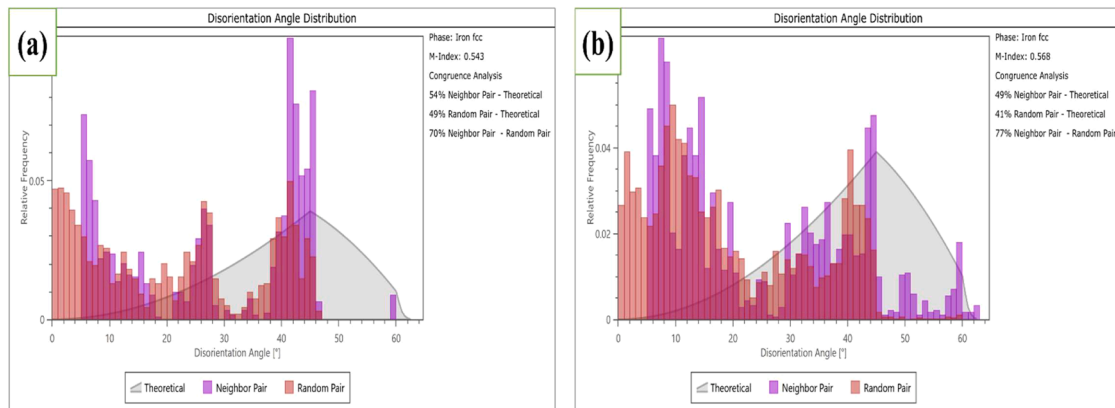


Fig. 13. Grain disorientation angle distribution analysis of (a) linear pattern, (b) Weaving Pattern.

exposed conditions. The variations in YS and UTS were 10.26% and 9.05% in the linear and weaving patterns, respectively. However, the variations in YS and UTS were 1.46% and 5.12%, respectively, for the gaseous hydrogen-exposed condition. Based on the variation of the hydrogen-exposed condition, the weaving pattern is highly resistant to HE failure. The higher the percentage of YS and UTS variation, the greater the strain hardening and the greater the increase in the ductility of as-printed hydrogen-exposed samples. Meanwhile, the lower the YS-UTS variation, the lower the work hardening and the higher the plasticity. Materials with high strain-hardening behaviour absorb greater deformation and control crack initiation, thereby delaying crack propagation.

3.4. Residual stress performance

Additive manufacturing uses layer-by-layer metal deposition techniques to fabricate materials, in which the input material (wire) is melted and deposited in accordance with the design. However, layer-by-layer metal deposition increases heat accumulation, leading to an uneven, steep thermal gradient and residual stress within the material. Due to this type of stress, changes in material properties, such as temperature stability, phase transformations, and mechanical properties, may lead to internal or surface cracking, fatigue degradation, and material warping. Fig. 16 illustrates the non-hydrogen-exposed linear and weaving pattern of (a, b and e, f) Debye ring representation, (c and h) distortion pattern, and (d and i) tested sample. The residual compression stress of the linear pattern is -176 MPa, and the weaving pattern is -143 MPa. Fig. 17

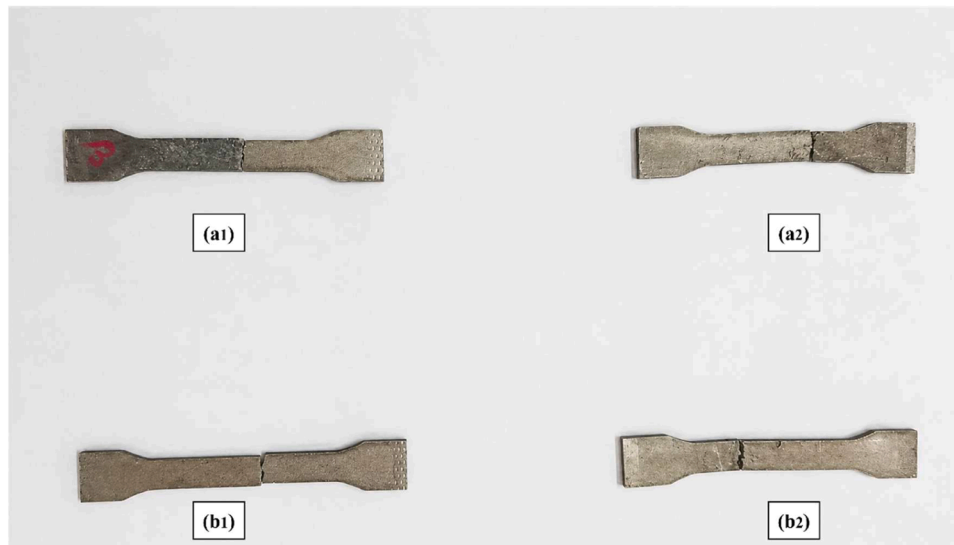


Fig. 14. Tensile test samples without and with exposure to gaseous hydrogen.

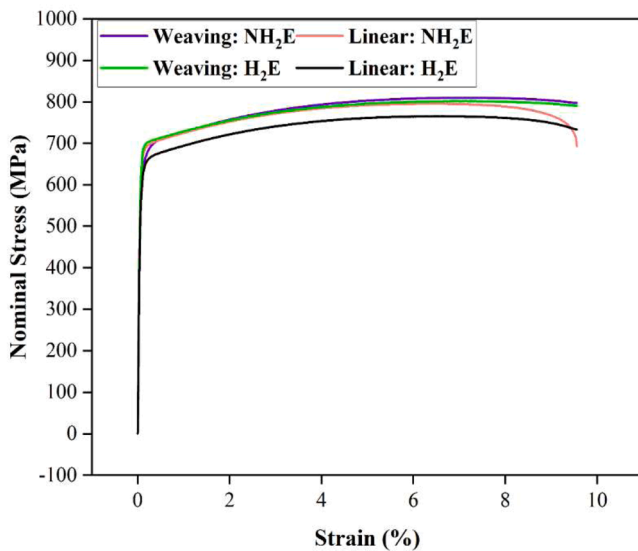


Fig. 15. Stress-strain diagram for WAAM-SS316L linear and weaving pattern samples for gaseous hydrogen-exposed and without exposure conditions.

illustrates the hydrogen-exposed linear and weaving pattern and their residual stress performance of (a, b and e, f) Debye ring representation, (c and h) distortion pattern and (d and i) tested sample. For the gaseous hydrogen-exposed condition, residual stress increased to -358 and -504 for linear- and weaving-pattern samples, respectively. Based on the distortion ring data compared to the weaving pattern, the linear pattern is highly distorted. Due to its high metal deposition and large molten pool formation, non-uniform heating and cooling, and greater thermal expansion and contraction result. In the linear pattern of the as-printed parts, the metal deposition follows localised heating, followed by rapid contraction.

In contrast to the linear deposition pattern, the weaving deposition approach uses an oscillating zigzag torch motion throughout the WAAM process to disperse heat across a larger area of the produced layer. Increased overlap between subsequent weld beads prolongs the thermal cycle and encourages a comparatively slower cooling rate, which reduces localised overheating and produces more even heat dissipation. As a result, the reduced temperature gradient promotes uniform solidification and grain evolution throughout the deposited material and limits

the development of residual stress.

In the gaseous hydrogen-exposed condition, the residual compression stress for the linear and weaving patterns is -504 MPa and -358 MPa, respectively. Compared with the linear pattern, the weaving pattern shows lower residual compression stress under both hydrogen-exposed and unexposed conditions.

3.5. Microhardness analysis of with and without hydrogen-exposed linear and weaving pattern

The materials are developed using CMT technology, with a strong focus on controlling microhardness. The lower heat input applied to developed WAAM components enables better control of molten-pool overflow and collapse. A high molten pool results in high angular distortion in as-printed parts, leading to irregular microhardness development on their surfaces. Fig. 18 shows the microhardness graph for linear and weaving patterns under hydrogen-charging (H2E) and non-hydrogen-charging (NH2E) conditions. Based on 10 trials, CMT-based WAAM shows lower deviations, particularly in the weaving-based patterns, with only 3% deviation in both hydrogen-exposed and unexposed conditions. Due to the low-heat-supply concept adopted in the CMT technology, the process doesn't experience molten-pool overflow, resulting in reduced anisotropy. Based on the results, the average microhardness of the linear pattern without and with hydrogen exposure is 186 HV and 191 HV, respectively, whereas for hydrogen-exposed weaving, it is 210 HV and 213 HV, respectively. However, the microstructure consists of a combination of austenite and ferrite phases. The austenite phase has high ductility, whereas the ferrite phase is highly brittle. For this reason, deviations are present in the as-printed samples.

4. Conclusion

The present work evaluated the influence of linear and weaving deposition patterns on the microstructure, mechanical properties, and hydrogen embrittlement behavior of WAAM-produced AISI 316L stainless steel before and after gaseous hydrogen charging. The following conclusions are drawn from the experimental results and comprehensive microstructural analyses.

- In comparison to many other metal additive manufacturing methods, the WAAM-CMT process offers better arc stability and a reduced heat input. By minimising thermal fluctuations during deposition, the decreased thermal input suppresses secondary or carbide phase

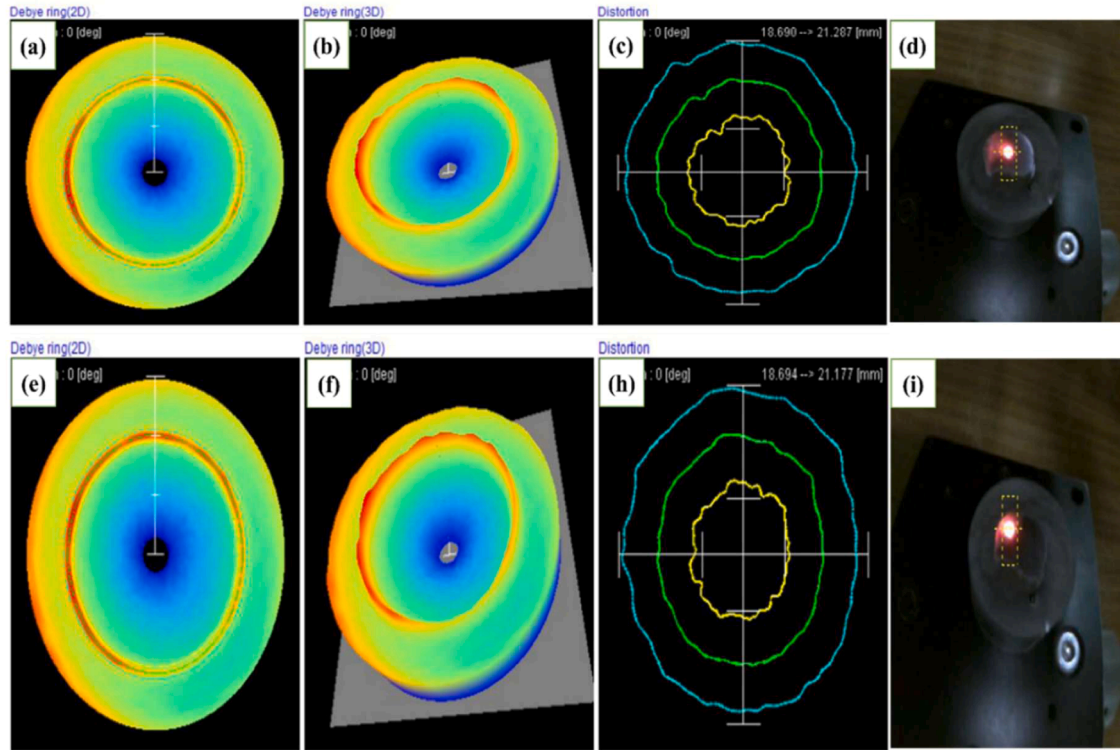


Fig. 16. Non-hydrogen-exposed linear and weaving pattern of (a, b and e, f) Debye ring representation, (c and h) distortion pattern and (d and i) tested sample.

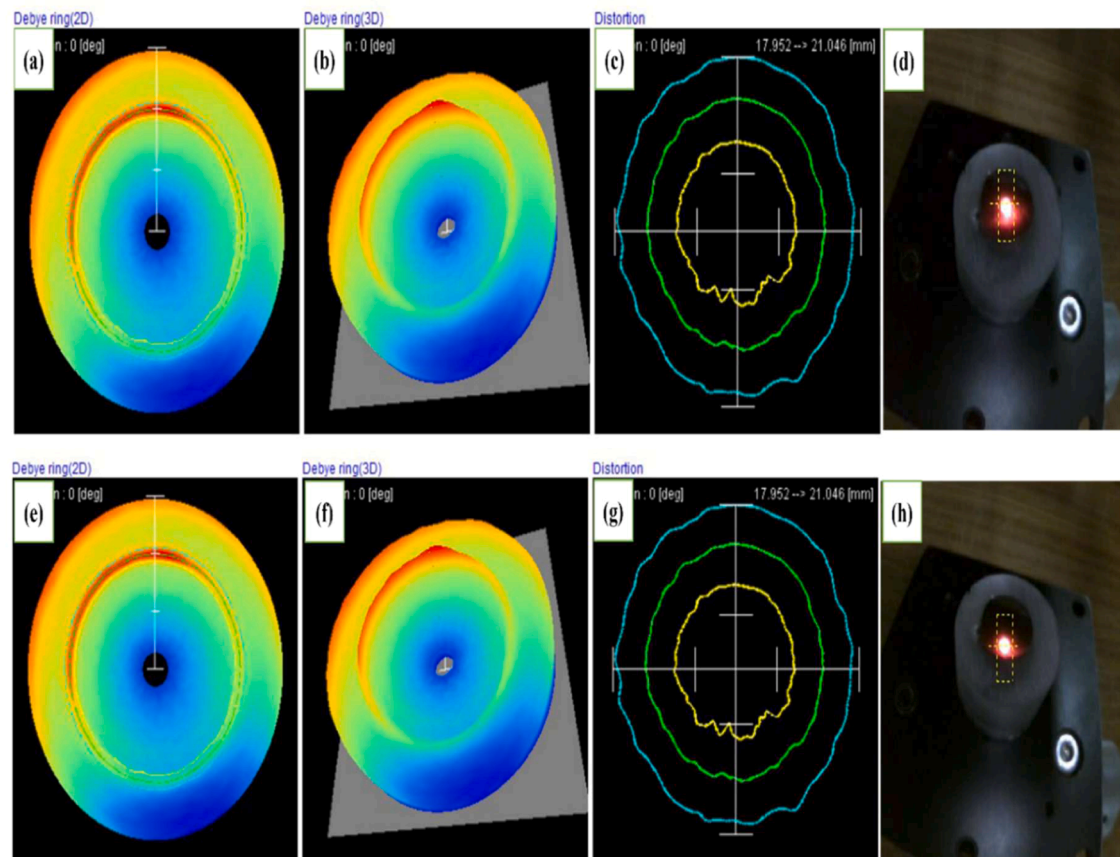


Fig. 17. Gaseous-hydrogen-exposed linear and weaving pattern of (a, b and e, f) Debye ring representation, (c and h) distortion pattern and (d and i) tested sample.

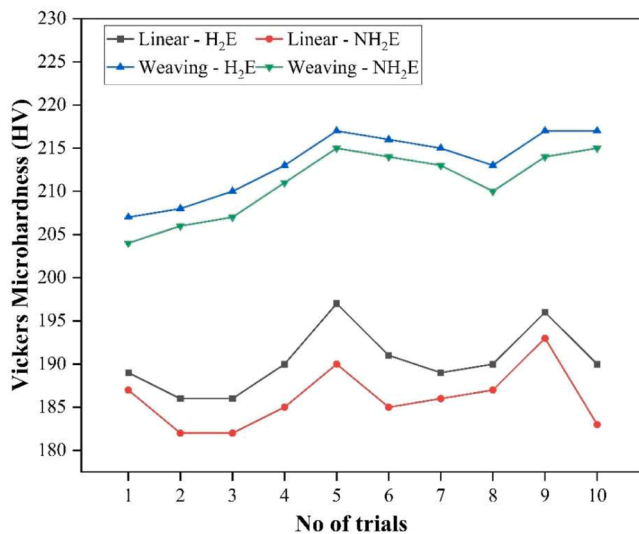


Fig. 18. Microhardness representation of linear and weaving patterns with and without hydrogen exposure conditions.

transitions and encourages the creation and maintenance of a stable FCC (austenitic) microstructure, with a minor amount of ferrite phase present.

- X-ray diffraction analysis confirmed the retention of the FCC (austenitic) phase in both deposition patterns, indicating that the low heat input associated with the WAAM-CMT process effectively stabilized the austenitic microstructure. Furthermore, the weaving deposition pattern exhibited reduced diffraction peak broadening compared with the linear pattern, suggesting improved crystallinity and lower lattice distortion.
- Elemental characterization by EDS revealed that the deposited material preserved the chemical composition of the SS316L feed wire. Compared with the linear deposition strategy, the weaving pattern showed better agreement with the nominal feed-wire composition, indicating a more uniform elemental distribution and reduced segregation during solidification.
- EBSD characterization demonstrated that the weaving deposition strategy developed a more stable microstructure with lower lattice distortion, reduced intragranular strain, and a well-defined crystallographic texture, contributing to enhanced resistance to hydrogen-assisted deformation and embrittlement.
- The stress-strain analysis demonstrated the elastic and plastic deformation behaviour of the WAAM-fabricated AISI 316L specimens under both hydrogen-exposed and uncharged conditions. The weaving deposition strategy exhibited superior tensile performance by maintaining higher strength and ductility after hydrogen exposure compared with the linear deposition strategy. These results indicate that the weaving pattern is more resistant to hydrogen-assisted degradation and exhibits improved resistance to hydrogen embrittlement.
- The WAAM-CMT process produced a stable austenitic microstructure with relatively low hardness, which is beneficial for reducing hydrogen embrittlement susceptibility. Although hydrogen exposure caused a slight increase in hardness due to lattice distortion and hydrogen-induced strain hardening, both deposition patterns retained good microstructural stability, with the weaving pattern exhibiting superior resistance to hydrogen-assisted degradation.
- Based on the perspectives compared to the linear pattern, the weaving pattern has less strain development and higher resistance to HE. Materials with high strain highly promote HE failures.

CRediT authorship contribution statement

Balaji V: Investigation, Formal analysis, Data curation. **Jeyapandiarajan P:** Resources, Methodology. **Ashwath P:** Supervision, Project administration. **Andre Batako:** Visualization, Supervision. **Sudhagar P:** Visualization. **Anthony Xavier M:** Writing – review & editing, Validation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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