

On the composition gradient of steel/Invar functionally graded material manufactured by wire-based direct energy deposition

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ARTICLE INFO

Keywords:

Composition gradient
Functionally graded material
Wire-based direct energy deposition
Microstructure evolution
Properties

ABSTRACT

This study utilized double-wire plasma arc direct energy deposition to produce functionally graded materials (FGMs) with two transition designs, abrupt (AT) and gradual (GT), from Er90s steel to Invar. The study systematically compared the transition in chemical composition, microstructure, phase evolution, thermal stress, and mechanical performance. Both FGM types exhibited a band structure in the Er90s section and coarse columnar grains in the Invar section, with the AT deposit showing a 1 mm thick, defect-free interface and the GT deposit having an 18 mm thick transition region with distinct boundaries. It revealed diverse microstructures across the transition zones, including fine ferrite, martensite with minor retained austenite (RA), coarse columnar austenite with martensite dendrites, and single FCC austenite. The GT sample uniquely featured a microstructure of martensite laths inside prior austenite decorated by RA semicontinuous network, with a crack detected due to dilatational stresses from martensite transformation. Hardness was similar in both FGM types, with higher values at the interfaces, especially in the GT FGM. The GT FGM demonstrated higher strength but lower ductility compared to the AT FGM, with failure occurring in the Invar portion for both. Thermal stress modelling indicated smoother stress transitions in the GT sample but no significant performance differences between Er90s and Invar. This study showcases the effectiveness of double-wire plasma arc DED in producing steel/Invar FGMs with varying composition gradients. It also underscores the importance of selecting the right mixing ratio for Er90s/Invar FGM deposits to avoid cracking and deterioration of properties in the gradient area.

1. Introduction

Additive manufacturing has emerged as a transformative technology with far-reaching implications across various sectors, including aerospace, automotive and maritime engineering, etc, due to its unparalleled design flexibility, enhanced material efficiency, and reduced need for expensive tooling and machining [1,2]. Wire-based Direct Energy Deposition (W-DED) holds immense promise for industrial applications due to its capability to manufacture large-scale components with complex geometries in open working environments [3]. W-DED has been extensively used to deposit complex geometric structures using a wide range of metallic materials (including aluminium, titanium, nickel-based superalloy, steel, etc.). So far, there has been less interest in developing functionally graded materials (FGM) components. W-DED offers a potential groundbreaking capability in the realm of graded metal material component manufacturing. The use of multiple wires

simultaneously fed into the molten pool via a multi-wire system integrated with W-DED allows for the seamless integration of multiple metallic materials within a single component, enabling the production of FGM. This facilitates the tailoring of distinct properties (mechanical, thermal, electric, or corrosion-resistant) across different regions of a component, thereby optimising its performance for specific applications [4,5]. For instance, a part could be engineered to have high tensile strength in one area and superior thermal resistance in another, all within the same manufacturing cycle. This capability opens up new avenues for innovation in sectors like aerospace, automotive, and energy, where materials with spatially varying properties can offer significant advantages in terms of both performance and cost-efficiency.

In an effort to lower carbon emissions and develop more fuel-efficient aircraft, the aerospace sector is increasingly incorporating lightweight composite materials like Carbon Fibre Reinforced Polymer (CFRP). During the moulding of such composites, differences in the

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<https://doi.org/10.1016/j.addma.2024.104594>

Received 12 September 2024; Received in revised form 19 November 2024; Accepted 6 December 2024

Available online 7 December 2024

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coefficients of thermal expansion (CTEs) between the resins, fibres, and mould materials can result in geometric distortions and interfacial stresses within the final component [6]. The CTE of CFRP typically ranges from 2.9 to 3.6 $\mu\text{m}/\text{m}\cdot\text{K}$, considerably lower than most metallic alloys [7]. Invar alloy (namely FeNi36), known for its low thermal expansion between 1.2 and 2.7 $\mu\text{m}/\text{m}\cdot\text{K}$ especially in the temperature range of 20°C and 200°C due to a balance of thermal expansion and magnetostriction effects and stabilised in its ferromagnetic phase [8], emerges as an ideal material for moulds used in CFRP manufacturing. It allows for the preservation of tight dimensional tolerances during temperature-variable forming processes. As a result, Invar moulds induce significantly less geometric distortion compared to those made from tool steel. This dimensional stability, coupled with surface quality and durability, has contributed to its widespread use and lasting appeal in the production of composite moulds [9,10]. Although Invar meets many of the requirements of an ideal tooling material, the alloy is relatively expensive compared to other tooling alloys (approximately 16 times the cost of carbon steel) [11]. For cost-saving purposes, a multi-material additive manufacturing process is a promising technique for reducing the amount of Invar needed while maintaining required thermal properties. FGM Parts can be manufactured using W-DED with an Invar profile and a steel substructure (such as low-carbon steel) to tailor the thermal expansion properties to specific applications while enhancing the economic feasibility of Invar fabrication.

Despite these advantages of applying steel-Invar FGM in moulds for CFRP production, deposition strategy development, especially in the steel-to-Invar transition, is a critical task. The disparity in the CTEs between Invar and steel can result in significant strain mismatches and elevated residual stresses, potentially causing defects such as cracking [12]. Additionally, the diffusion of elements can give rise to unwanted phases [13], particularly in the transition zone between steel and Invar, making this area crucial to the performance of the whole FGM parts. Depending on application needs and material variances, the transition in FGM can be either abrupt or gradual, each with its own set of advantages and drawbacks. Abrupt transitions may lead to heightened stresses and chemical incompatibility, gradual transitions could mitigate these issues. However, mixing multiple materials in a single melt pool may result in the formation of undesirable intermetallic compounds, compromising weldability and processability, consequently, increasing the risk of premature failure [5]. Arbogast et al. [11] produced a steel-Invar FGM using laser DED by directly depositing Invar on top of a steel substrate and evaluated the composition and CTE transition across the interface. The results demonstrate that additively manufactured Invar can effectively lower the CTE, making it a promising material for multi-material part tooling applications. However, the deposition rate of laser DED is constrained by the laser's energy output and the precision required for melting the wire feedstock. In contrast, wire-based DED (WDED) offers significant advantages for large-scale structure manufacture, including very high build rates, 100 % material utilisation, high-quality feedstock, defect-free deposition, unlimited build volume, and most importantly, low cost [14].

In this study, steel-Invar FGM single-pass wall structures were deposited using plasma-transferred arc-based (PTA) WDED with different transition gradients. The effect of the transition gradient from steel to Invar on the evolution of mechanical performance, microstructure and CTE was systematically evaluated. This investigation provides a theoretical basis and valuable reference for using W-DED to manufacture low-cost and high-performance moulds for CFRP production.

2. Experimental

2.1. Material and WAAM setup

The feedstock utilized in this investigation consisted of Er90s steel and Invar wires with a diameter of 1.2 mm. S355 steel plates with dimensions of 300 mm $\times$ 200 mm $\times$ 12 mm were employed as the

substrates. The chemical compositions of these materials are shown in Table 1. The experimental arrangement for the PTA WDED process is shown in Fig. 1(a). The entire setup was enclosed and filled with pure argon gas, maintaining the oxygen level below 800 ppm. The PTA was generated using an EWM power source. The positioning of each wire feeder nozzle relative to the substrate surface was set at an angle of 35°, and the two wire-feeding nozzles are positioned at an approximate angle of 45° to each other (Fig. 1 (b1-b2)). A CMOS camera (Xiris XVC-1000) was positioned beside it to observe the metal transfer behaviour into the melt pool. The substrate was fixed to a workbench using clamps.

To evaluate the influence of composition transition mode from Er90s to Invar on the process, deposit morphology, microstructure and mechanical performance, two ways (gradual transition-GT and abrupt transition-AT) were selected to deposit the multi-layer single-pass Er90s-Invar FGM wall structures, with key process parameters listed in Table 2. For AT, the first 40 layers were solely Er90s and layers 41–98 were deposited using Invar. For GT, Er90s wire and Invar wire were simultaneously fed into the melt pool, with their constituent ratio controlled by their individual wire feed speeds (WFS), as indicated in Table 2. Travel speed (TS) was fixed at 4.5 mm/s during the whole deposition process. In this case, by gradually reducing the WFS of Er90s and increasing the WFS of Invar, the gradual transition from Er90s to Invar could be achieved while the deposition rate remains unchanged. The mass fraction (E_x) of alloying elements in the deposits can be calculated using the following equation [15]:

$$E_x = \frac{\sum WFS_i \rho_i E_{xi}}{\sum WFS_i \rho_i} \quad (1)$$

where E_{xi} ($x = \text{Fe, Ni, Nb, et al.}; i = 1, 2$) is the mass fraction of a certain element in a distinct wire, WFS_i ($i = 1, 2$) is in m/min, D_i ($i = 1, 2$) is the wire diameter in mm, ρ_i ($i = 1, 2$) is the density of the wire in g/cm³. The predicted chemical compositions in wt% with varied WFS of Er90s and Invar are calculated accordingly and listed in Table 3.

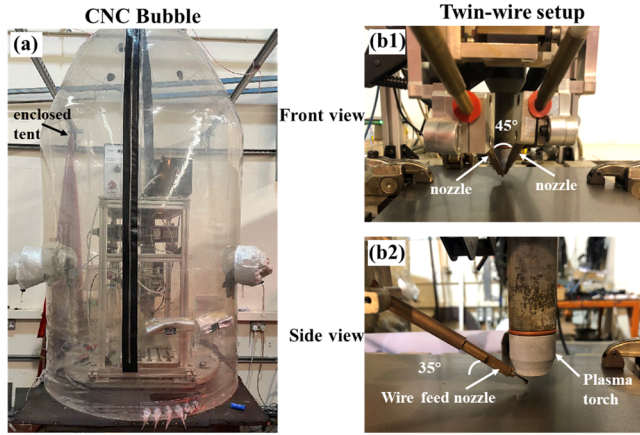
2.2. Characterisation

A 5 mm thick section was taken from the building direction (BD)-normal direction (ND) plane in the middle of both FGM structures for microstructural, compositional and hardness analysis. These samples were ground, polished, and etched using a 5 % Nital solution to reveal the microstructure. A field emission gun scanning electron microscope (SEM, TESCAN S8000G) with Energy-dispersive X-ray spectroscopy (EDS) and electron backscattered diffraction (EBSD) detectors was used to study the microstructure and crystallographic orientation across the transitional areas. EBSD scanning settings included an accelerating voltage of 15 kV, a probe current of 6.5 nA, and a step size of 0.5 μm . The indexing rate of all EBSD maps conducted is above 88 %, showing good quality data. The collected EDS and EBSD data were post-analysed using Aztec and AztecCrystal software, respectively. Microhardness tests were conducted along the central line of the metallographic samples from steel to Invar using a Zwick/Roell Hardness Tester, applying a 300 g load and 0.5 mm spacing. The microstructures around indentations with typical hardness values were examined by optical microscope. Tensile tests were performed on flat specimens with gauge length of 25 mm, cut from the FGM components in the BD-transverse direction (TD) plane with gauge region covering composition transition, in accordance with ASTM E8 standards, at room temperature and a constant strain rate of 1 mm/min. Three tensile samples were taken from each wall structure for data reliability, and the fractured surfaces were subsequently examined using SEM. To measure variation in the CTE with mixing ratio, six single-pass walls with a fixed chemical composition corresponding to the designed mixing ratio were deposited. Cylinder specimens with dimensions of ϕ 4 mm $\times$ 5 mm were extracted from each wall in the vertical and horizontal planes. The CTE tests were carried out on a TMA450 thermomechanical analyzer, with samples

Table 1

Chemical composition in wt% of filler wires and substrate.

Materials	C	Si	Mn	Cr	Mo	Ni	V	Cu	Nb	Fe
Er90s steel Wire	0.09	0.56	0.90	2.5	1.06	< 0.1	< 0.01	< 0.1	< 0.01	Bal
Invar Wire	0.2	-	0.4	-	-	36	-	-	1.6	Bal
S355 Substrate	≤ 0.20	≤ 0.55	≤ 1.60	-	-	-	-	≤ 0.55	-	Bal

**Fig. 1.** Experiment setup: (a) overview of the CNC bubble setup; (b1) front view and (b2) side view of the configuration of the plasma torch, double wires, and substrate.**Table 2**

Key deposition parameters for different transition modes.

Transition mode	layer	Current (A)	WFS _{Er90s} (m/min)	WFS _{Invar} (m/min)
Gradual transition—GT	1–30	220→200	2.1	0
	31–35	190	1.5	0.6
	36–40	190	1.2	0.9
	41–45	190	0.9	1.2
	46–50	190	0.6	1.5
	51–98	190→170	0	2.1
Abrupt transition—AT	1–40	220→200	2.1	0
	41–98	200→170	0	2.1

heated up to 800°C at a heating rate of 10 °C/min, held for 30 mins and then cooled down to room temperature.

3. Finite element thermal modelling

FE models were developed to simulate stress distribution across the transition regions of the two FGMs at elevated temperatures referring to the operating temperature of the autoclaves. As shown in Fig. 2, two FE models with dimensions of 50 mm × 100 mm × 3.75 mm were created. The element type adopted in these two models is C3D8T element, which is a coupled displacement-temperature 8 nodes brick solid element with Gauss integration points. The coupled element allows the FE solver to

compute thermal and mechanical results at the same time and save computational time. Two material property arrangements were designed for each of the FE models to simulate two FGM structures with either a GT arrangement or an AT arrangement between the Er90s regions (in yellow) and Invar regions (in green). The GT region was divided into four sections with heights of 5 mm according to the composition gradient, as shown in Fig. 2(a). The AT region was divided into two sections with heights of 1 mm, as shown in Fig. 2(b). The mesh was locally refined at the transition boundaries to capture the stress and strain peaks due to property mismatch. A minimum element size of 0.2 mm was adopted. The quantity of finite elements in AT FGM is 17200, and in GT FGM is 23200.

X-symmetry boundary condition was applied to both horizontal side edges of the single-pass wall structures. One point node on the base was fixed in the y-axis displacement and rotation while one point node on the top was fixed in the x-axis displacement and rotation to prevent the drifting of the model. To simulate the autoclave loading conditions, an external pressure load of 1.2 MPa was applied on all free surfaces, but not the X-symmetry boundary surfaces. The initial temperature for all the nodes was 25°C. Three operating temperatures (200°C, 300°C and 400°C) were simulated. Temperature-dependent material properties, including CTE, thermal conductivity, and Young's modulus for different regions corresponding to different mixing ratios, were also applied to the two models. Thermal conductivity and Young's modulus values were interpolated according to the material ratio between steel and invar at the graded regions, shown in Fig. 3(a) and (b), respectively. Both are found to be temperature-dependent and composition-dependent. From room temperature to 400°C, both thermal conductivity and Young's modulus decrease with the increasing proportion of Invar. But the differential narrows as the temperature rises.

4. Results

4.1. Melt pool

Fig. 4 compares the deposition processes with decreasing WFS of Er90s but increasing WFS of Invar. For stable deposition, the droplet surface tension transfer mode is maintained where the droplet contact with the melt pool is approximately equivalent to the time required to grow a new droplet regardless of the varying WFS ratio. It has been reported that such a transfer mode could mitigate the risk of spattering and maintain a consistent frequency of droplets [16,17]. In the case of constant transfer mode, the melt pool shape changes accordingly with the change of Invar proportion. With the increment of Invar addition, the periphery of the melt pool expands gradually, indicating an increasing melt pool size. This is because the thermal conductivity of

Table 3

Predicted chemical composition in wt% with varying WFSs of Er90s and Invar.

Condition	WFS (m/min)		Predicted Chemical composition (wt%)								
	Er90s	Invar	Ni	Fe	C	Mn	Nb	Si	Cr	Mo	Cu
W1	2.1	0	0	96.35	0.1	0.8	0	0.4	1.6	1	0.15
W2	1.5	0.6	10.40	86.08	0.13	0.68	0.46	0.28	1.14	0.71	0.11
W3	1.2	0.9	15.57	81.18	0.14	0.62	0.69	0.23	0.91	0.57	0.09
W4	0.9	1.2	20.71	76.30	0.16	0.57	0.92	0.17	0.68	0.42	0.06
W5	0.6	1.5	25.83	71.44	0.17	0.51	1.15	0.11	0.45	0.28	0.04
W6	0	2.1	36	61.8	0.2	0.4	1.6	0	0	0	0

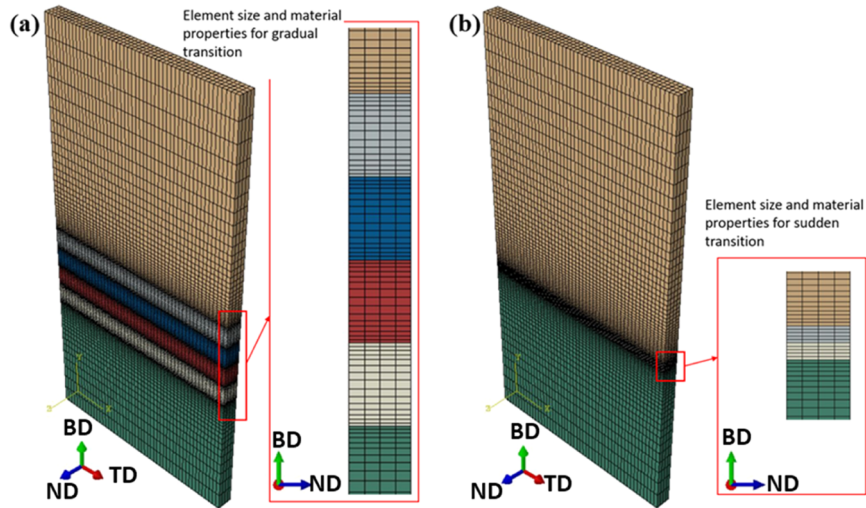


Fig. 2. Material property variations with varying composition in the transition region of (a) GT (b) AT FGM represented by the varying colours and transition boundaries showing reduced finite element size.

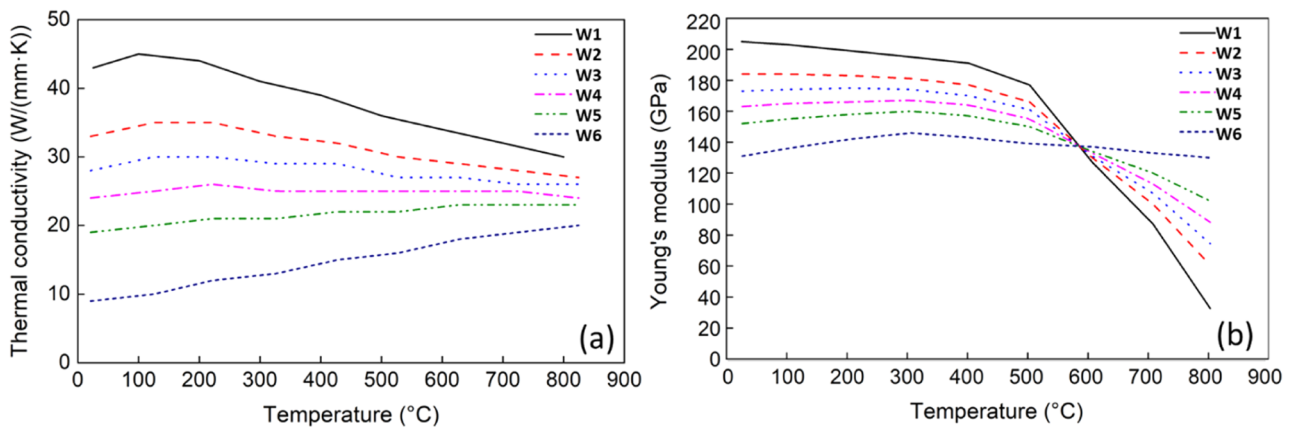


Fig. 3. Temperature-dependent material properties: (a) thermal conductivities and (b) Young's modulus.

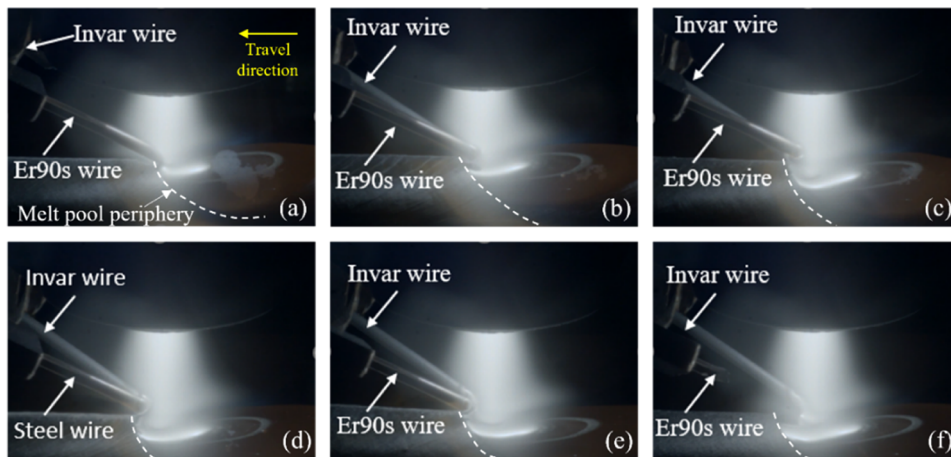


Fig. 4. Deposition process of different conditions with varied Er90s/Invar wire feed speed ratio: (a) W1 – 0 wt% of Ni; (b) W2 – 10.4 wt% of Ni; (c) W3 – 15.57 wt% of Ni; (d) W4 – 20.71 wt% of Ni; (e) W5 – 25.83 wt% of Ni; (f) W6 – 36 wt% of Ni.

Invar is quite low compared with Er90s steel (Invar: 10.5 W/m·°C [18]; Er90s: 39 W/m·°C [19]), leading to a lower heat transfer rate, therefore, a slower cooling rate together with a prolonged solidification process.

Moreover, due to its very low CTE, Invar experiences minimal solidification shrinkage during cooling. As a result, the melt pool of Invar tends to exhibit reduced contraction and deformation compared to Er90s steel

which undergoes more significant solidification shrinkage.

4.2. Coefficient of thermal expansion

Fig. 5 shows the CTE variation as a function of temperature at different mixing ratios. CTE is not highly sensitive to temperature in the compositions of W1-W5, as shown in Fig. 3(a-e). In contrast, both W6 samples extracted from horizontal and vertical planes display notably low thermal expansion strain ($<4 \times 10^{-6}$ mm/°C) spanning from room temperature up to about 230°C, as shown in Fig. 3(f). This 'Invar effect' is attributed to a balance between thermal expansion and magnetic ordering changes in the alloy, leading to minimal net expansion. Once this critical temperature threshold is surpassed, there is a sharp increase in thermal expansion strain, leading to linear thermal expansions. Research [20] suggests that this shift to linear thermal expansion could be attributed to atomic vibrations at 230 °C, which effectively counteract the magnetovolume phenomenon. By plotting CTE as a function of Ni content at different temperatures, as shown in Fig. 6, there is an overall downward trend in CTE values with the increment of Invar proportion, but not a linear decrease in the temperature range of 100°C - 400°C. Ni stabilizes the austenitic phase with a densely packed structure that expands less when heated compared to BCC structures, as it allows for greater absorption of thermal energy without significant expansion. Additionally, Ni strengthens metallic bonds within the steel, making the atomic structure more resistant to thermal expansion. This stabilization of the microstructure, combined with the homogeneity and solid solution strengthening effects of Ni, results in a lower CTE, making Ni-rich steels less susceptible to dimensional changes with temperature fluctuations [21].

4.3. Macro morphology and composition transition

Etched cross-sectional macrographs of Er90s-Invar FGM deposits with different transition modes are shown in Fig. 7. In both AT and GT conditions, the band structure in light contrast that curves downward can be clearly observed. In WAAM deposits, it was reported [22] that

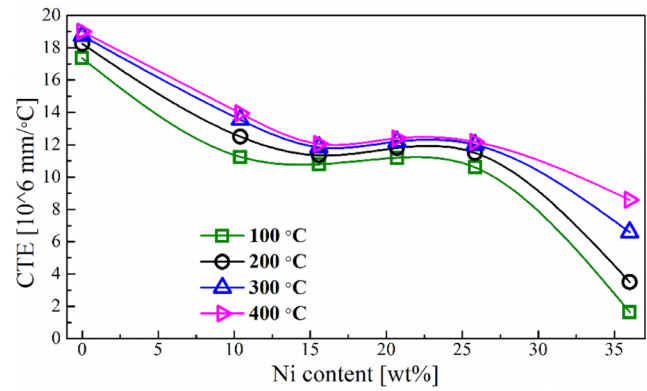


Fig. 6. CTE variation with Ni content at different temperatures.

this etching contrast is caused by subtle fusion boundary segregation. Coarse columnar grains in mm scale were found to extend through multiple layers and grow epitaxially along BD in the Invar portion since grain formation is favoured along the direction of maximum heat flow [23]. As depicted in Fig. 7(a), the interface region of the AT deposit is approximately 1 mm thick and free of macro defects. In contrast, the transition region of the GT deposit measures around 18 mm in thickness, and distinct interface boundaries with both the Er90s and Invar sections are visible (Fig. 7(b)). Fig. 8 illustrates the chemical composition transition along the BD in the transition region from position A of GT, as determined by SEM-EDS analysis at 0.5 mm intervals. To ensure data reliability, a $500 \mu\text{m} \times 50 \mu\text{m}$ (width $\times$ height) area was chosen for composition measurement at each location, rather than a single point. Although the design called for an incremental addition of Invar every five layers in a stepwise manner, the actual deposition process resulted in a relatively continuous transition with minor fluctuations in chemical composition. This was due to the remelting and reheating steps involved in the deposition. Each new layer underwent remelting, affecting the upper part of the preceding layers, while the lower part experienced

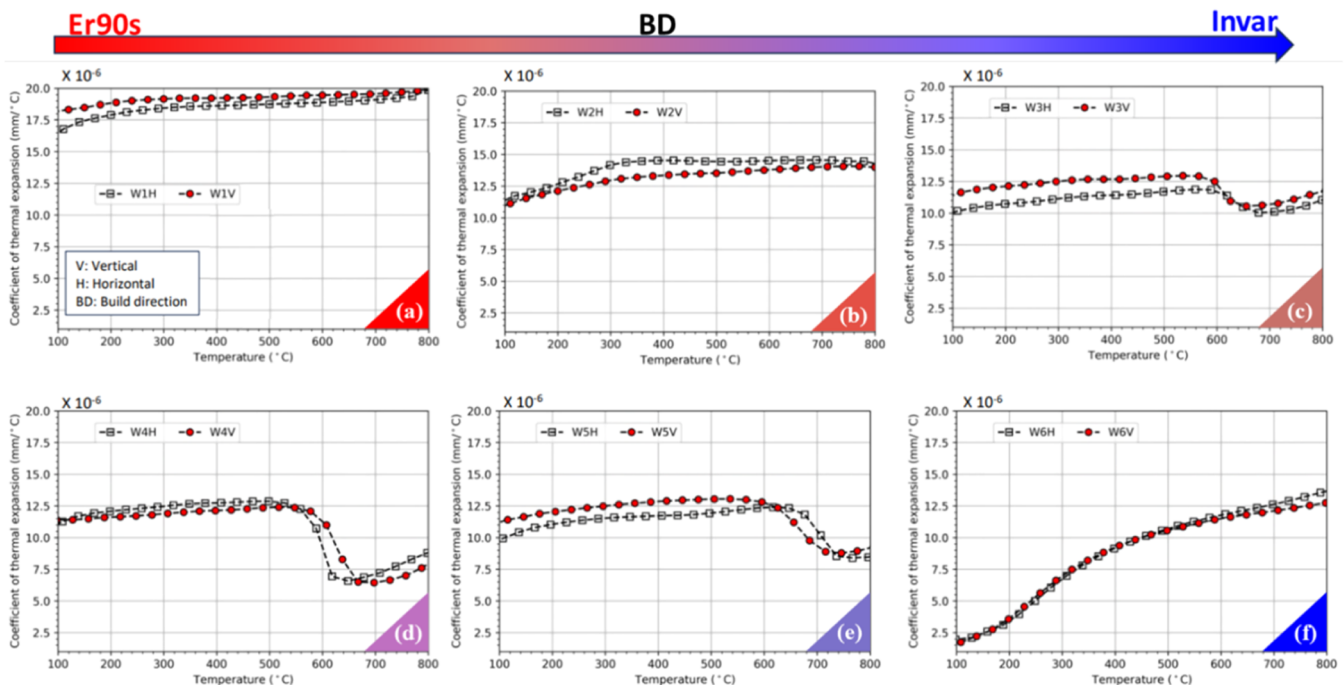


Fig. 5. Coefficient of thermal expansion (CTE) as a function of temperature with varying chemical composition along the BD in the interface region of GT FGM: (a) W1 – 0 wt% of Ni; (b) W2 – 10.4 wt% of Ni; (c) W3 – 15.57 wt% of Ni; (d) W4 – 20.71 wt% of Ni; (e) W5 – 25.83 wt% of Ni; (f) W6 – 36 wt% of Ni. H – samples are extracted from the horizontal plane; V – samples are extracted from the vertical plane.

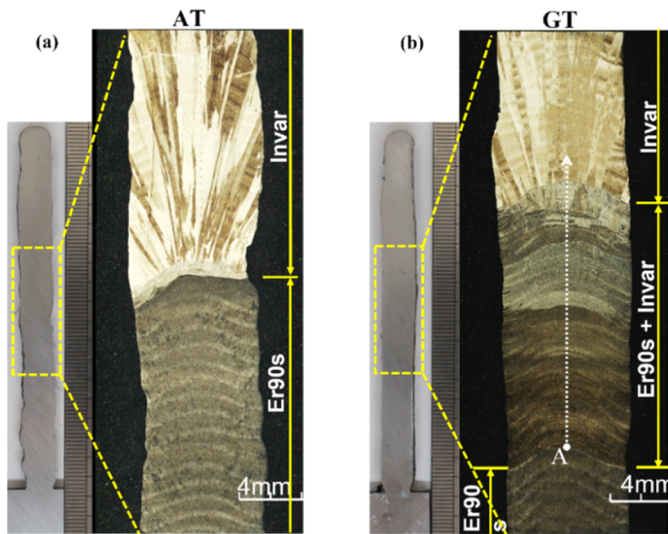


Fig. 7. Cross-sectional macro morphologies of Er90s-Invar FGM deposited by different transition modes: (a) abrupt transition—AT; (b) gradual transition—GT.

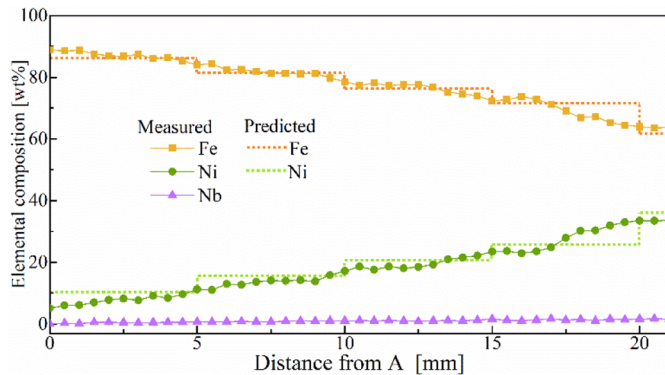


Fig. 8. Chemical composition transition along the BD in the interface region from position A of Er90s-Invar FGM deposited by GT mode.

reheating and a secondary solid-phase transition [24]. This process also moderated the abrupt composition gradient of Fe and Ni, leading to a smoother transition rather than a stepwise one.

4.4. Thermal stress analysis

Fig. 9 compares the Von Mises stress distribution across the transition region of the Er90s-Invar FGM with GT and AT transition modes at different operation temperatures. The resultant value of the thermal expansion coefficient in the graded region was directly dependent on the material ratio. A higher CTE would result in higher stress in a restricted condition. As a result, the stress was presented in stairway-decreasing distributions across the transition regions in both the GT and AT samples that match the decreasing trend of the CTE. Increasing the operating temperature from 200°C to 300°C shows an increase in the stress magnitude in the steel region. Further increase in temperature from 300°C to 400°C, however, reduced the stress magnitude in the steel region instead. This could be because the CTE of steel as a function of temperature increases from 200°C to 300°C, then remains constant from 300°C to 400°C, while Young's modulus of steel decreases when temperature increases from 200°C to 400°C. The stress magnitude of the invar region, however, showed a constant increase with the temperature from 200°C to 400°C. This could be explained by the fact the CTE of Invar increases steeply with temperature while Young's modulus of

Invar remains relatively constant. However, despite the difference in the slope of the stress transition between the AT and GT, the magnitude of stress within the steel regions and the invar regions are similar between the two models under all working temperatures. The only effect is that the transition region was extended, and the stress transition changed from a sharp drop to a gradual descending. In both AT and GT arrangements, no sign of concentration of stresses was shown. This is evidence that the arrangement of the transition region did not affect the performance of either the steel region or the invar region under the practical working conditions.

4.5. Tensile performance

The tensile properties of Er90s-Invar FGMs in the case of AT and GT are shown in Fig. 10 and Table 4. The average ultimate tensile strength (UTS) and yield strength (YS) of the GT FGM are relatively higher when compared with those of the AT FGM. However, this advantage in strength comes at the expense of more than half ductility (AT: 16.4 %; GT: 7.4 %). Both FGMs are almost 1.5 times the UTS in comparison to pure Invar deposited by WDED [25], but the ductility is much lower. However, it can be found that the ductility of AT FGM is comparable to that of Er90s deposited by WDED [26]. Fractured tensile specimens, as shown in Fig. 11, confirm that the failure of both FGMs has occurred in the Invar portion, while the interface just went through subtle deformation. This indicates that lower YS and higher ductility enable the Invar portion to deform initially when subjected to external forces when compared with Er90s and transition region in the FGM. From the fractured specimens depicted in Fig. 11(a1, b1), obvious deformation can also be observed outside the gauge region. Especially, the fracture location of the GT FGM specimens is almost at the edge of the gauge region and very close to the reduced section, as can be seen from Fig. 11(b1), indicating that the real ductility should be much larger than the tested ductility. The gauge length of the tensile specimens is determined to be 25 mm according to the ASTM E8 standard, which could cover the transition region of both FGMs. However, the area fraction of invar in the gauge region of GT specimens is much smaller than that of AT ones. Accordingly, the contribution of Invar to elongation (EL) in GT specimens is much smaller, so the measured EL is much smaller than that of the AT specimens. Therefore, the ductility value here is closely related to the length and position of the gauge taken and only for reference. The fracture morphologies of GT and AT samples look very similar to each other, showing the presence of dimples and secondary cracks, which is evidence of a ductile failure induced by void nucleation, growth, and coalescence. Secondary cracks often act as stress concentrators, diverting stress away from the primary crack tip. This could slow down the propagation of the main crack during the tension process, thereby enhancing the fracture resistance of the material [27]. Overall, the secondary cracks in the fracture surface of the AT samples are smaller in size and more evenly distributed, thereby playing a more significant role in delaying the onset of fracture.

4.6. Hardness transition and typical microstructure evolution

The microhardness variations across the transition region of Er90s-Invar FGMs under AT and GT conditions are shown in Fig. 12(a) and Fig. 13(a), respectively. The hardness of the Er90s part and Invar part is similar in both AT and GT conditions: 250–330 HV in Er90s while 160–175 HV in Invar. The scattering in hardness value is much higher in Er90s than in Invar. This is because the indents are occasionally made on or near the bands, and randomly chosen test positions may indent on the different phases, causing variation in the hardness for the adjacent two positions. Corresponding microstructure around indents with typical values are shown in Fig. 12(b1–b6) and Fig. 13(b1–b6). Er90s portions are mainly composed of ferrite, bainite and martensite, as depicted in Fig. 12(b1) and Fig. 13(b1), which is a typical microstructure of low-carbon low-alloy steel [26]. Invar portions are found to be dominated

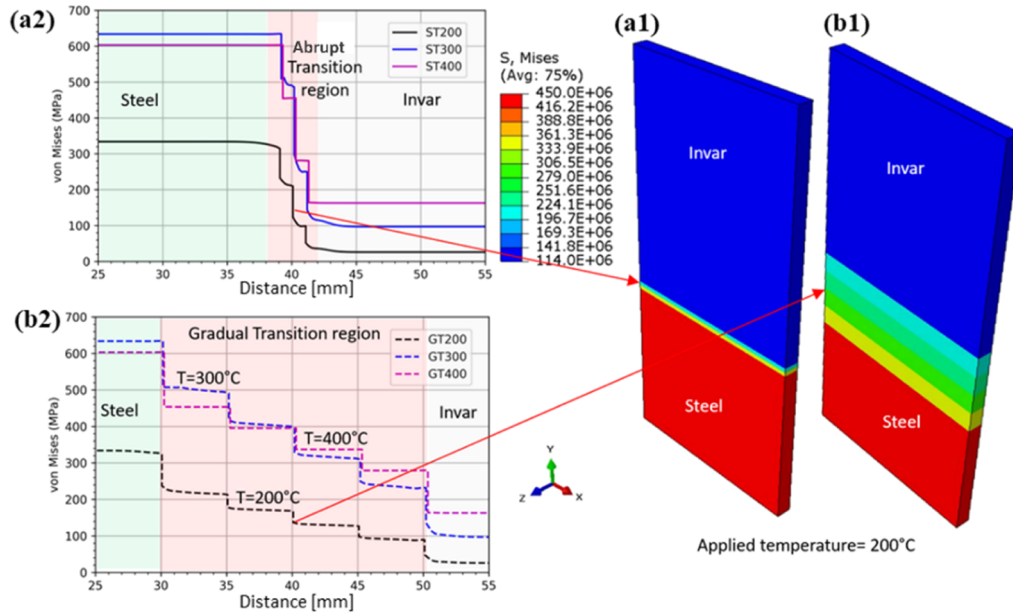


Fig. 9. Von Mises stress distribution in the transition region at different operation temperatures: (a1-a2) AT; (b1-b2) GT.

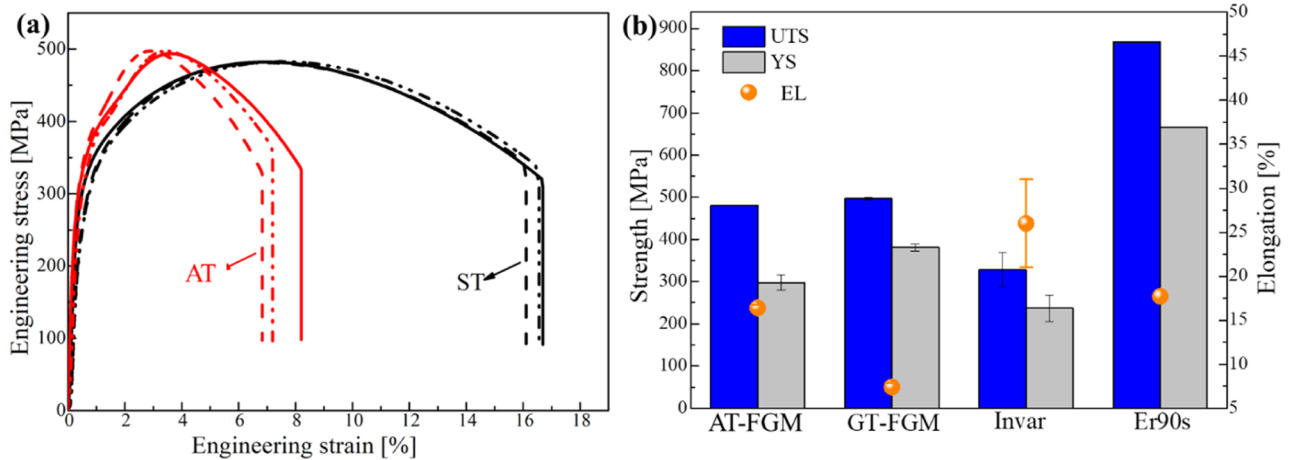


Fig. 10. Tensile properties of Er90s-Invar FGM deposited by different transition modes (a) engineering strain-stress curves of AT and GT specimens; (b) comparison of the corresponding ultimate tensile strength (UTS), yield strength (YS) and elongation (EL) of AT-FGM, GT-FGM, pure Invar and pure Er90s deposited by WDED.

Table 4

Tensile properties comparison of AT-FGM, GT-FGM, pure Invar and pure Er90s deposited by WDED.

Material	UTS, MPa	YS, MPa	EL, %
AT Er90s-Invar FGM [This study]	481.2 ± 0.7	298 ± 18.5	16.4 ± 0.4
GT Er90s-Invar FGM [This study]	496.7 ± 2.9	381 ± 8.9	7.4 ± 0.8
Invar [25]	329 ± 41	237 ± 31	26 ± 5
Er90s steel [26]	869	667	17.7

by columnar grains with columnar or cellular dendritic substructure (Fig. 12(b5, b6) and Fig. 13(b6)). These distinct substructures are typically observed in single-phase austenitic alloys, developed due to localized shifts in the solidification conditions within the melt pool and influenced by both the temperature gradient and growth rate at the solid-liquid interface [28]. In the AT FGM sample, the hardness value at the interface (position C) is found to be higher than the surroundings. The microstructure at this position (Fig. 12(b4)) is composed of martensite and retained austenite dual phases due to the increased Ni

content from the dilution effect of Invar, responsible for the abrupt increase in hardness.

A hardening zone above 350 HV can be found in the transition region of the GT FGM sample, which could be attributed to the Ni content variation [29]. Position A, with Ni content of approximately 5.1 wt%, depicts a transition from a combination of ferrite + bainite + martensite to predominant martensite showing coarse columnar prior austenite (PA) morphology with dark contrast at boundaries, which is similar to that of 5Ni martensitic steel reported by [30]. It was reported that once Ni content has exceeded 4 wt%, PA will be distinctly coarsened with enrichments by Ni, Mn and Mo at interdendritic regions [29]. With Ni content increased to 8.4 wt%, the matrix of Position B is still martensite with very little retained austenite (RA), which is analogous to 9Ni low-carbon martensitic steel reported by [31]. From position B to D, the hardness value gradually decreases from 420 HV to around 350 HV with increased Ni content to 18.6 wt%. The matrix remains to be martensite but the RA content is gradually increased and mainly rich at the prior austenite grain boundaries, which is the main reason for the hardness reduction. In position E, with Ni content determined to be 21.5 wt%, a

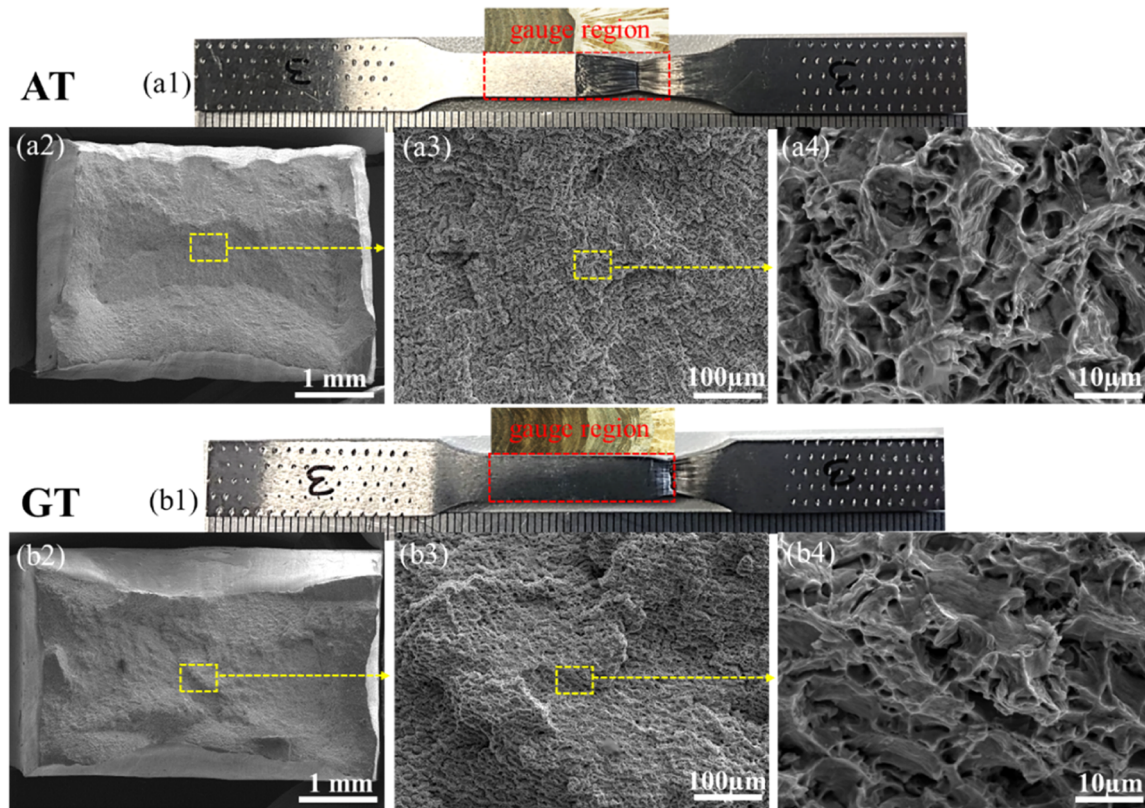


Fig. 11. Fracture morphologies of Er90s-Invar FGM deposited by different transition modes (a1-a4) AT; (b1-b4) GT.

distinct transition from a martensite matrix with RA secondary phase to an austenite matrix with martensitic dendrites distributed can be observed, resulting in a significant drop in hardness (Fig. 13(b5)). Further increase in Ni leads to a decreasing fraction of martensite and getting closer to the typical microstructure of Invar, as can be seen in Fig. 12(b6). It is known that Ni addition in steel helps to stabilize the RA and prevent the transformation of austenite to martensite upon cooling to room temperature [32]. With increasing Ni content, on one hand, the austenite phase field shifts towards lower temperatures, on the other hand, the solubility of carbon in austenite increases, which further stabilises the austenitic phase [33].

4.7. Evolution of grain orientation, phase, and dislocation

To evaluate the influence of composition transition on the microstructural evolution of Er90s-Invar FGMs, EBSD was performed on the transition zone processed with AT and GT conditions, viewed in the BD-ND plane. The grain orientation (IPF, inverse pole figure), phase distribution and geometrically necessary dislocations (GND) density in the Er90s/Invar interface of the AT FGM sample are shown in Fig. 14 (a), (b), and (c), respectively. As indicated, visually identifiable regions across the Er90s/Invar interface could be found in terms of grain morphology and phase composition. Along BD, four zones can be distinguished: zone I is mainly composed of fine ferrite with an average size of 4.3 μm and an aspect ratio of 1.9, which is typical grain morphology of Er90s; zone II consists of lath martensitic BCC and minor RA FCC dual phases, with a mean grain size of 8.6 μm and an aspect ratio of 2.8; zone III is identified by coarse columnar austenite FCC grains with martensite BCC dendrites; zone IV is single FCC phase zone made up of coarse austenite columnar grains in mm scales, which is typical structure of Invar. The IPF map indicates that the austenite grains grew across multiple deposited layers in regions III and IV, with no discernible fusion lines. Moreover, the absence of a dendritic structure in region IV,

as evidenced by the IPF map, suggests that the grain orientations in both the interdendritic and dendritic zones are closely aligned. The austenite grains with the same orientation extended from region III to IV, indicating epitaxial growth of the austenite phase in different materials. From the GND density map shown in Fig. 14(c), the highest GND density could be found in Zone II while the lowest GND density turned out to be zone IV. This is because martensite in zone II is formed from the diffusionless transformation that essentially freezes the atoms in a highly strained configuration, leading to a significant increase in dislocation density [34]. In contrast, ferrite in zone I and austenite in zone IV form through diffusional transformations, which allow atoms to move and rearrange into low-energy configurations, minimizing the number of dislocations. Both austenite (FCC structure) and ferrite (BCC structure) are thermodynamically more stable and less strained than the body-centred tetragonal (BCT) structure of martensite [35].

As the transition region of the GT FGM sample is designed to be very wide, in accordance with hardness value guidelines, six distinct zones have been strategically selected within the extensive transition zone of the GT sample for a comprehensive EBSD analysis. Corresponding orientation maps and phase maps are shown in Fig. 15 and Fig. 16, respectively. The microstructure in region a is mainly composed of the BCC phase, showing the morphology of major lath martensite and a few bainite and ferrites in the coarse columnar original structure. With layers added up, the microstructure turned into the lath martensite phase while the columnar original structure became finer (region B). The average proportion of RA increased to 5.2 %, mainly distributed at the grain boundary of the original austenite columnar along BD. When transiting to region C, the RA content further increased to approximately 17 %, forming a semicontinuous network structure. With the gradual increase of Ni content, austenite structure grows from the original grain boundary to the grain interior with the proportion further increased to 28 % in region D. When reaching region E, the austenite network structure became continuous, and the morphology turned into cellular

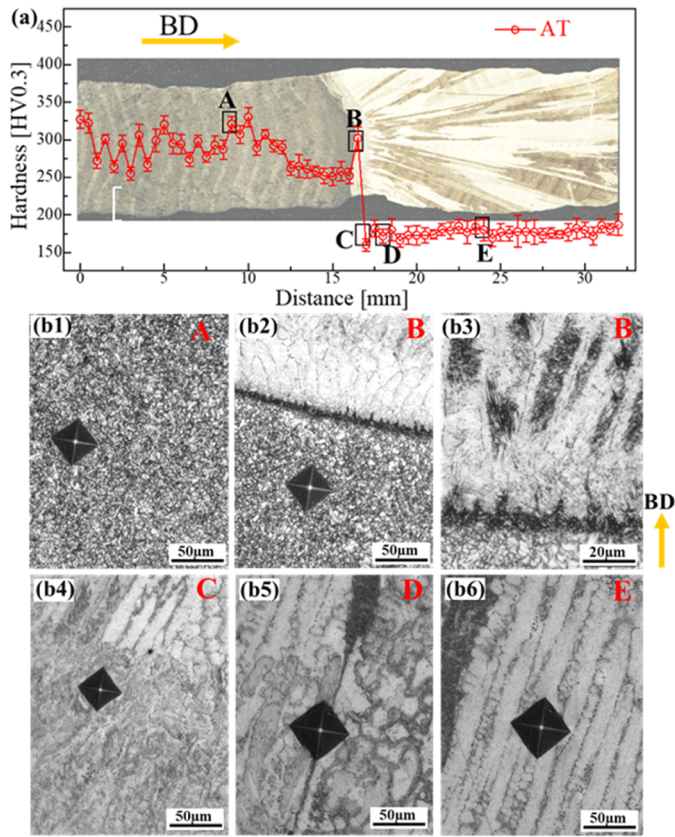


Fig. 12. (a) Hardness transition along the BD across the interface region of Er90s-Invar FGM deposited by AT mode; (b1-b7) microstructures around the indents at positions A-E accordingly.

with martensite laths inside distributed on the austenite matrix. Such cellular morphology gradually decreased to disappear with the stacking of layers, as can be seen in region F. The austenite phase close to the Invar section exhibited pronounced (101) and (100) orientation, a result of epitaxial growth. This suggests that the austenite phase formed directly from the liquid phase [36,37]. It should be noted that the microstructure of martensite laths and RA semicontinuous network shown in regions C and D only appears in the GT sample while being absent in the AT sample.

To understand the evolution of the plastic activity in the different zones across the transition region for the GT sample, the GND maps are calculated and plotted in Fig. 17. In general, the warm area expanded from region a to b, indicating that the average GND density rises, with peaks at approximately $7 \times 10^{14} \text{ m}^{-2}$ inside the martensitic blocks. The cool colour region is mainly distributed on the prior austenite boundaries. With layers added up, the cool colour area becomes wider, implying the average GND density gradually decreases. Even so, the distribution of GND density is not uniform, still showing high values varying from $4 \times 10^{14} \text{ m}^{-2}$ to $7 \times 10^{14} \text{ m}^{-2}$ in the lath martensite cellular. In region f, there is a distinct reduction in GND density to $0.15 \times 10^{14} \text{ m}^{-2}$ could be observed when the microstructure transits to a single FCC structure. Typically, a high GND density leads to elevated levels of total dislocation energy in specific local areas. This makes it challenging for dislocations to pass through grain boundaries, thereby inhibiting grain boundary migration and plastic deformation [38]. As a result, material strength tends to increase significantly in regions a-c. Conversely, a low GND density facilitates slip transmission and plastic deformation in other areas, particularly in regions e and f.

One thing worth mentioning is that a crack with a length of around 100 μm existed in region c in the GT sample, as can be clearly seen in Fig. 18. This is an intergranular crack, located at the retained austenite

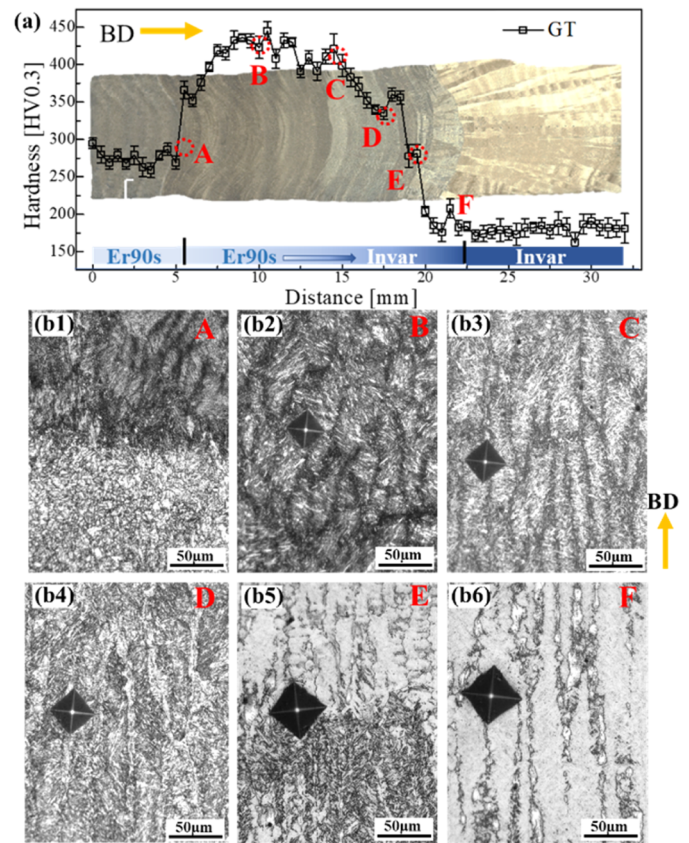


Fig. 13. (a) Hardness transition along the BD across the interface region of Er90s-Invar FGM deposited by GT mode; (b1-b7) microstructures around the indents at positions A-F accordingly.

region and propagates along the grain growth direction. From the phase and GND density distribution, the crack tips are in the FCC region, which is the weak zone and much softer (lower GND density) compared with the BCC martensite blocks. The difference in mechanical properties between martensite and austenite can lead to stress concentration and localized deformation. When subjected to external loads or thermal cycles, these stress concentrations can exceed the material's fracture toughness, leading to crack initiation and propagation. Also, during cooling from the austenitizing temperature, the austenite phase transforms into martensite, leading to volume expansion and the generation of residual stresses. These stresses can be concentrated at the interfaces between the martensite and austenite phases, making these areas more susceptible to crack initiation. Therefore, this crack can be determined to be a cold crack, forms due to stress concentration at the interface between the retained austenite and martensite phases, which often occurs after the material cools down, particularly when the austenite transforms to martensite. Furthermore, the presence of impurities, inclusions, or pre-existing micro-cracks can act as stress concentrators, accelerating the crack formation process. In some cases, the austenite phase may also undergo a partial transformation to untempered martensite during deformation, which is brittle and can facilitate crack formation. Therefore, the formation mechanism of cracks here could be a multifaceted issue involving metallurgical, mechanical, and environmental factors. There are also several precautions for such cracking formation. First, it's essential to manage cooling rate through proper preheating or interpass temperature control to prevent cold cracks during deposition. Second, controlling process parameters (such as energy input and material input) could help minimise residual stresses and martensite formation. Most importantly, chemical composition range with high cold crack susceptibility could be minimised or even avoided

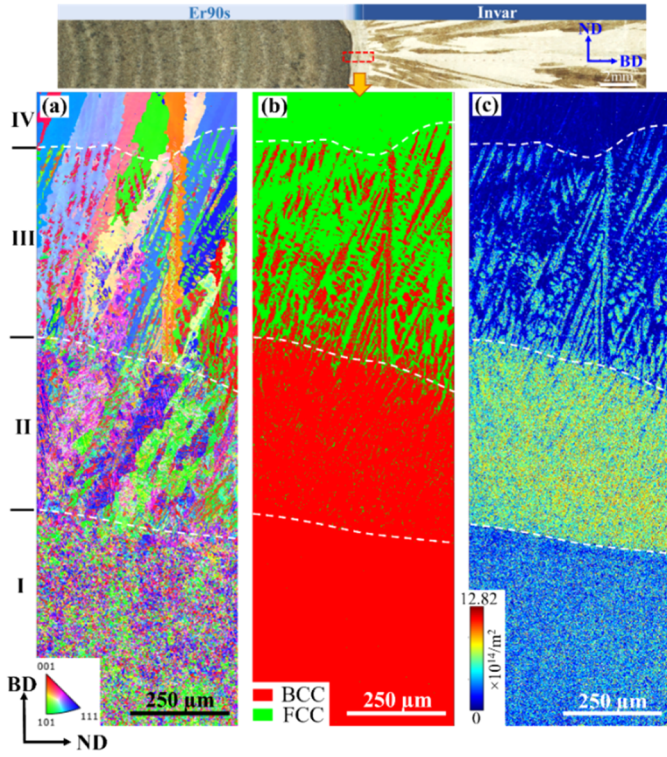


Fig. 14. EBSD analysis on region across the interface of AT Er90s-Invar FGM: (a) IPF map; (b) phases distribution map presenting the BCC in red and FCC in blue; (c) GND density map.

by adjusting the WFS ratio in double-wire plasma arc direct energy deposition process.

5. Discussion

The microstructural evolution in the transition region from Er90s to Invar in the AT sample exhibits distinct characteristics compared to the

GT sample, which could be attributed to the deviation in the chemical composition resulting from the predetermined mixing ratio and the inherent effects of remelting and reheating during the WDED process. In the AT sample, Invar is deposited directly onto the Er90s layer, leading to a sharp transition in composition and microstructure, significantly influenced by localised remelting and reheating. The most notable chemical shift is observed in the Ni content, which escalates swiftly from under 0.1 wt% to approximately 36 wt% within a narrow transition zone of less than 1 mm. In contrast, in the GT sample, manual adjustments in the mixing ratio produce a broader transition zone of approximately 19 mm between Er90s and Invar. This comparison underscores the critical impact of Ni variations on microstructural evolution, highlighting its role in shaping microstructural characteristics.

Due to the similarity in principal properties, Ni and Fe form a substitutional solid solution when mixed. In the high-temperature range, the Fe–Ni binary alloy comprises a peritectic reaction such as the Fe–C binary alloy. Solidification from the liquid phase can proceed along various phase transformation scenarios:

For compositions with lower Ni additions (less than 3.4 wt%), and correspondingly low carbon content, solidification begins entirely as δ -ferrite, which subsequently transforms into austenite. This transformation concludes with the nucleation of α -ferrite along austenite grain boundaries, followed by its progressive growth [39]. Nickel alloying significantly expands the austenite phase field, and its low diffusivity within the austenite lattice delays the austenite-to-ferrite transformation to lower temperatures or longer times. This undercooling effect increases the ferrite nucleation rate while reducing its growth rate, resulting in a finer ferrite microstructure [40]. Additionally, the nucleation and growth of ferrite tend to obscure the prior austenite grain boundaries (PAGBs) [39]. Also, the formation of acicular ferrite and martensite required a threshold level of Ni. It is reported that acicular ferrite begins to form with increasing Ni content in an Fe–0.04 %C–0.7 %Mn alloy, and that approximately 30 vol% martensite appeared at 5.5 % Ni [41]. In this investigation, the Er90s zone in both AT and GT conditions shows a fine microstructure of ferrite, bainite, and martensite due to the combined effects of low nickel concentration and rapid cooling rates. As Ni concentration rises in the transition region (e.g. from zone I to zone II in AT sample; from Er90s zone to zone a in GT sample), martensite becomes more pronounced,

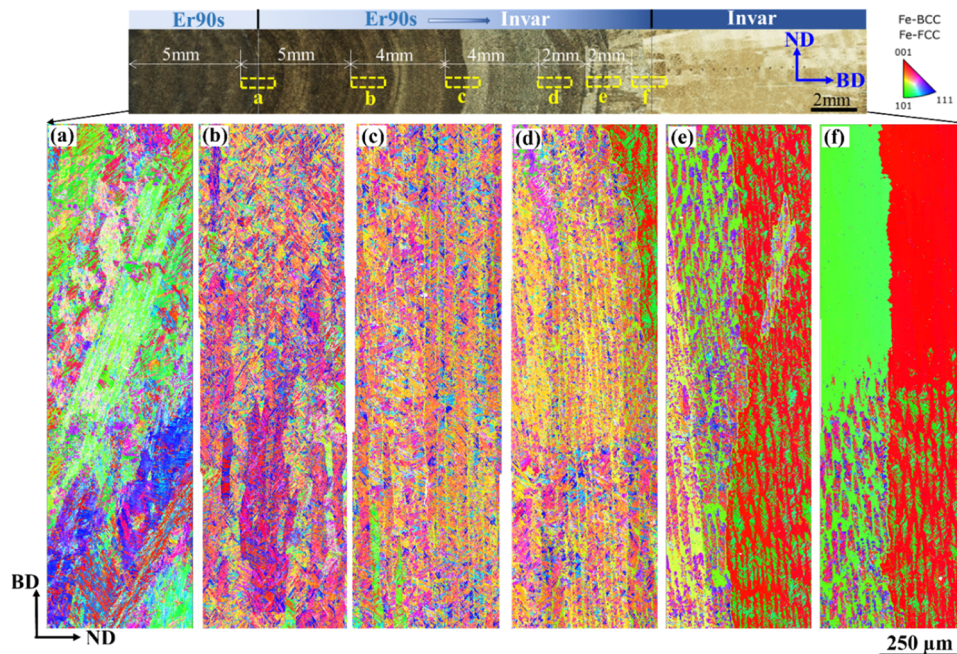


Fig. 15. IPF maps taken at different regions in the transition region of GT Er90s-Invar FGM.

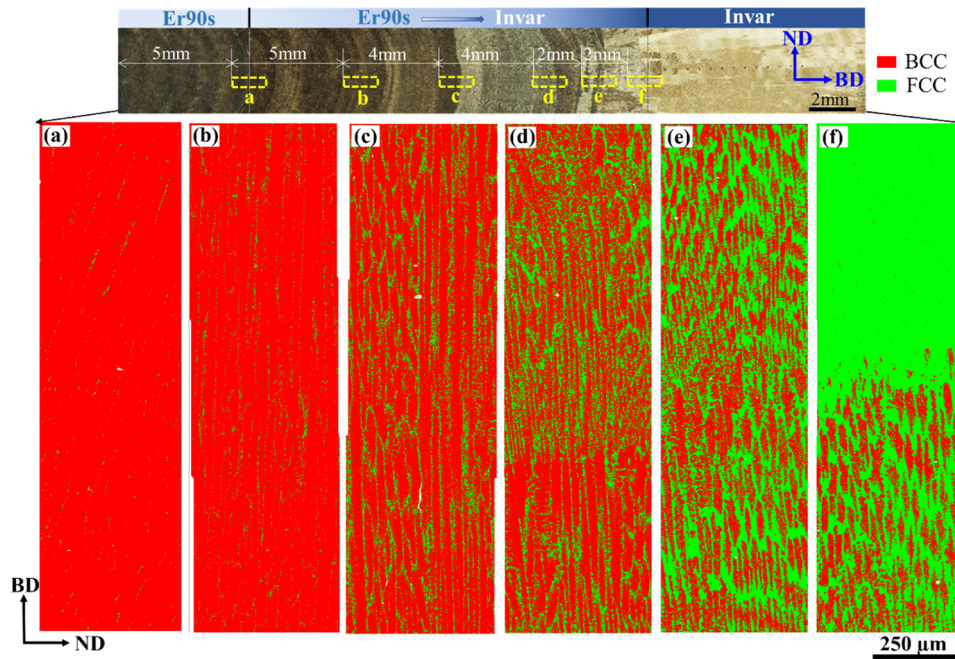


Fig. 16. Phase maps taken at different regions in the transition region of GT Er90s-Invar FGM.

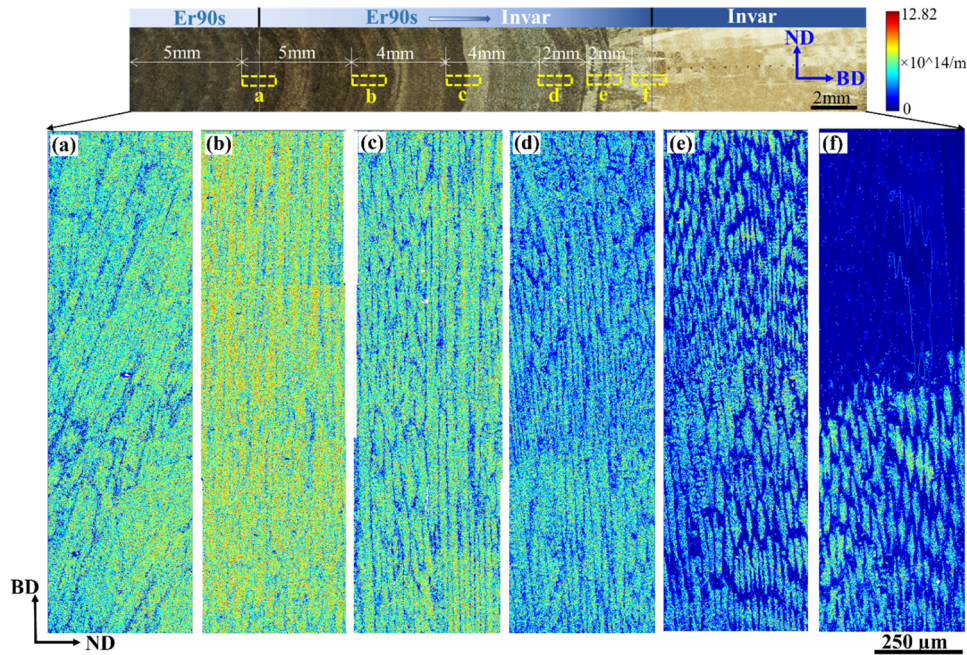


Fig. 17. GND maps taken from different regions in the transition region of GT Er90s-Invar FGM.

reflecting the progressive influence of Ni on microstructural evolution.

When the Ni content exceeds 6 wt%, diffusion-controlled transformation processes are entirely inhibited, leading to a martensitic transformation (liquid $\rightarrow$ liquid + austenite $\rightarrow$ austenite $\rightarrow$ austenite + martensite $\rightarrow$ martensite) [40,42]. The PAGBs are preserved, with the martensite substructure developing within these confines. Notably, after direct solidification from liquid to austenite, impurities such as S and P elements segregate to the PAGBs, thereby marking the PAGBs in the final martensitic microstructure [43]. Such morphology can be clearly observed in the transition region in both AT and GT conditions in this investigation. It is important to note that the austenite phase is present at room temperature when the nickel content is above 5 %, albeit in a

minimal proportion [40]. In such cases, the faster diffusion of nickel in Fe-BCC compared to Fe-FCC leads to a hysteresis effect in the transformation behaviour between cooling and heating cycles, becoming more pronounced. Martensitic transformation begins during cooling when the material passes the M_s (martensite-start) temperature. Before reaching the M_f (martensite-finish) temperature, both BCC and FCC phases coexist; however, the Ni concentration in each phase is nearly identical due to the extremely slow diffusion rate. Upon reheating above the A_s (austenite-start) temperature into the region where FCC and BCC phases coexist, Ni can partition into the austenite if the holding time is long enough. These Ni-enriched crystallites may ultimately persist as metastable retained austenite phases upon cooling back to ambient

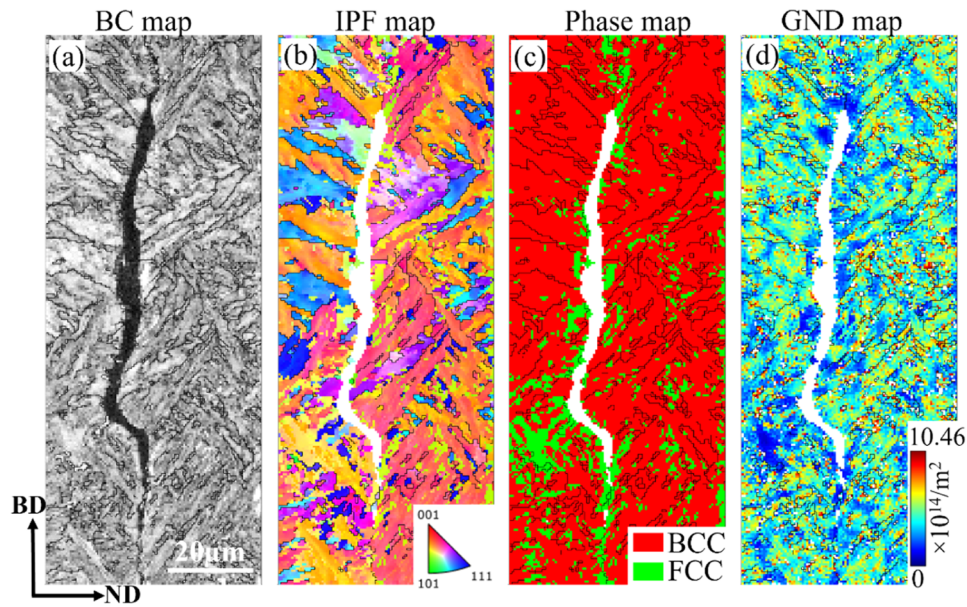


Fig. 18. Magnified EBSD maps of the crack appeared in region c: (a) band contrast (BC) map; (b) IPF map; (c) phase map; (d) GND map.

temperature [40]. In this study, zone II of the AT sample and regions a-c of the GT sample exhibit a similar martensitic microstructure with retained austenite structures distributed along the PAGBs. The formation of these retained austenite could be triggered by the reheating cycles during the deposition process. And it can be clearly observed in the transition region getting closer to the Invar region of both AT and GT conditions, the fraction of retained austenite increases due to the increasing Ni content.

When Ni content exceeds approximately 15 wt%, a dual-phase microstructure of ferrite and austenite becomes stable at room temperature. According to the Fe-Ni phase diagram [44], solidification begins with δ -ferrite formation, leveraging the stability of iron in a BCC structure at elevated temperatures. As cooling continues, δ -ferrite transforms into γ -austenite due to the influence of Ni, which reduces the transformation temperature and encourages the formation of FCC structure. At further reduced temperatures, the cooling rate plays a crucial role in determining the final microstructure: rapid cooling typically promotes martensitic transformation within austenite, while slower cooling stabilises retained austenite. During the deposition process, complex thermal cycles, such as melting, remelting, and reheating, produce a martensite-austenite dual-phase with a coarse columnar structure. Higher Ni content lowers the transformation temperature, favouring the formation of bainite or martensite upon cooling. Simultaneously, austenite phase nucleation occurs along PAGBs and progressively expands along these boundaries [40]. As Ni concentration increases further, a segregated network of austenite around martensite blocks develops, which becomes more pronounced with the continuous rise in Ni content. This phenomenon is visible in zone III of the AT sample and regions e and f of the GT sample, where the martensite blocks are delineated by austenite boundaries, resulting in a structure that enhances both toughness and resistance to deformation.

The microstructure of high-alloyed steels can also be made out using the Schaeffler diagram, which employs Cr and Ni equivalents (Cr_{eq} ; Ni_{eq}). It allows for the identification of distinct microstructural domains based on calculated Cr_{eq} and Ni_{eq} values, facilitating the prediction of phases in accordance with the chemical composition. The calculation of Cr_{eq} and Ni_{eq} for the materials involved is determined by coefficients that reflect the influence of each element on the stability of austenitic or ferritic phases [45]:

$$Cr_{eq} = \%Cr + \%Mo + 1.5 * \%Si + 0.5 * \%Nb + 2.0 * \%Ti \quad (2)$$

$$Ni_{eq} = \%Ni + 30 * \%C + 0.5 * \%Mn + 30 * \%N \quad (3)$$

The derived Cr_{eq} and Ni_{eq} values corresponding to the nominal chemical composition of the mixing Er90s and Invar with different ratios are displayed in Table 5. The corresponding coordinates are marked in the Schaeffler diagram, shown in Fig. 19(a). Conforming to the experimental results, Er90s steel possesses a mixture of ferrite and martensite while Invar is composed of austenite. When the proportion of Invar accounts for 43 % in the mixing, the coordinate sits in the martensite-austenite dual structure region, indicating a martensite-austenite duplex microstructure could be achieved under such a mixing ratio condition. With the increasing addition of Invar, the coordinate moves upward further and across this dual-phase region to the single austenite region. In this study, due to the dilution and diffusion of elements caused by remelting and reheating, the duplex phase region will be wider than in theory.

The transition of phase fractions from Er90s to Invar in both AT and GT samples is illustrated in Fig. 19 (b) and (c). For the AT sample, within the transition zone, the FCC phase fraction gradually rises to about 10 % over approximately 350 μ m, followed by a sharp increase to 75 %, where it becomes the predominant phase across a width of 115 μ m. This fraction then steadily reaches 100 %, becoming the sole phase over a span of around 440 μ m. In the GT sample, the FCC phase initially increases slowly to 30 % over a length of 13.3 mm, and then significantly jumps to 100 % within a 4.8 mm width. Through comparison, a martensite-austenite duplex structure with austenite content of less than 30 % in the AT sample occupies a much narrower area of less than 400 μ m, which is only 1/32 of that in the GT sample. Regarding the martensite-austenite duplex structure, the distribution and proportion of phases play a crucial role in performance and cracking tendency.

Table 5

Calculated Cr_{eq} and Ni_{eq} values correspond to different Er90s-Invar mixing conditions.

Er90s-Invar mixing condition	Cr equiv.	Ni equiv.
pure Er90s	3.2	3.4
Er90s: Invar = 70 %: 30 %	1.49	14.64
Er90s: Invar = 57 %: 43 %	2.17	20.11
Er90s: Invar = 43 %: 57 %	1.82	25.8
Er90s: Invar = 30 %: 70 %	1.76	31.19
pure Invar	1.22	42.2

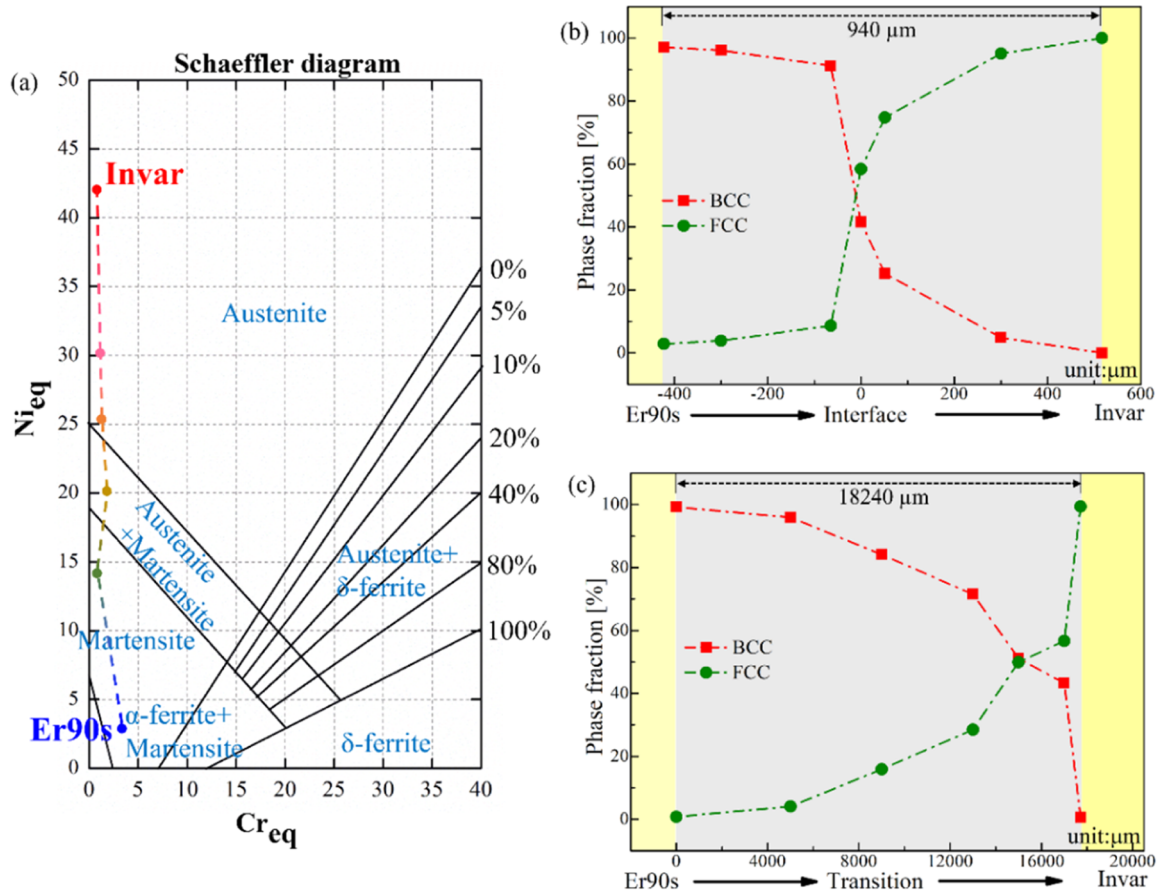


Fig. 19. (a) Microstructure corresponding to different mixing ratios is depicted in the Schaeffler diagram; phase fraction transition from Er90s to Invar (b) AT; (c) GT.

Melting of the Er90s and Invar wires occurred with the action of the arc, which was accompanied by remelting of the previously deposited layer. Upon cooling, the transformation into martensite introduces dilatational stresses that exert pressure on the PAGBs, where the RA is mainly distributed. This stress can localize in the softer RA, leading to void formation and embrittlement at these boundaries, potentially resulting in intergranular cracking, which is a critical concern for the performance of steels alloyed with high levels of Ni [46]. Even though the finer RA contributes to toughness, it still can act as a path for crack propagation in the form of a continuous network along PAGBs, which could explain the case that happened in the GT sample.

The evolution of chemical composition and microstructure in the transition zone also exert a remarkable influence in CTE. From this study, it can be found that CTE exhibited a nonlinear decreasing trend with the increase of Ni content. CTE is influenced by several factors related to atomic structure, composition, and environmental conditions. Low Ni content (< 10 wt%) would slightly raises CTE by stabilising the FCC austenitic phase and promoting moderate expansion due to retained BCC structures at certain levels, as FCC usually have higher CTE than BCC structures due to their atomic arrangement, which allows more room for thermal expansion. However, no increase in CTE was captured as Ni content increased in this study, which is because the Ni content of W2 is out of the 0–10 wt% range. When Ni content surpass 10 wt%, higher Ni levels lead to a substantial reduction in CTE, especially near the Invar composition (around 36 wt% Ni), where CTE is minimised. This low CTE results from restricted atomic movement due to stabilising effect of Ni on FCC phase and magnetic ordering, especially close to the Invar composition [47].

The evolution of chemical composition and microstructure in the transition zone also directly affects the hardenability. From the GND

density transition from Er90s to Invar of both samples shown in Fig. 20, the GND density first increases to $5\text{--}6 \times 10^{14} \text{ m}^{-2}$ and gradually decreases to less than $1 \times 10^{14} \text{ m}^{-2}$ with fluctuations in the transition regions. Such a trend is consistent with the hardness transition in both samples. It is evident that the hardenability increase is most prominent for smaller Ni additions, while at higher Ni content, the increasing effect on hardenability levels is off. A small addition of Ni lowers the critical cooling rate required for the formation of martensite, meaning that slower cooling rates can still produce a fully martensitic structure, contributing to higher hardenability. The hardness increase occurs along with a gradual shift from a ferritic towards a bainitic–martensitic microstructure and decreases with the increasing nucleation of austenite [48]. In both cases, the highest hardenability appeared in the transition region due to the massive formation of martensite, followed by the Er90s region, and the lowest was the Invar zone. This could also explain why all fractures occurred in the Invar region when tension stress was applied to them. The fact that the fracture occurred from the Invar side when subjected to tensile stress also provides integrity with the GND density and hardness values measured in these regions.

6. Conclusions

This study utilised double-wire plasma arc DED technology to deposit Er90s/Invar FGMs, comparing abrupt and gradual transition designs by controlling the dual-wire feeding speed ratio. The key findings are:

- (1) The AT formed a 1 mm defect-free interface, while the GT created a much wider transition region. In the GT sample, continuous chemical transitions and varied microstructures were observed,

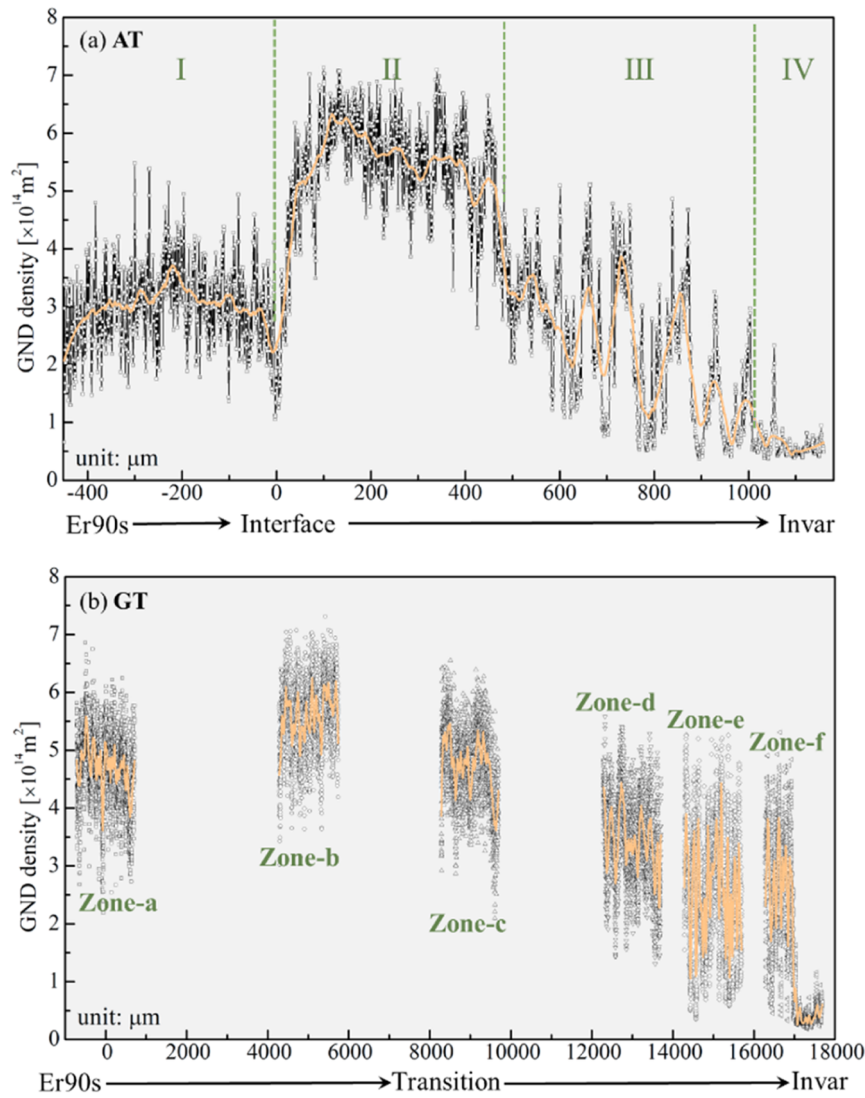


Fig. 20. GND density transition from Er90s to Invar (a) AT; (b) GT.

including austenite networks and martensite laths. This led to potential embrittlement, highlighting the importance of selecting the appropriate Er90s/Invar mixing ratio.

- (2) The CTE exhibited a nonlinear trend, with the Invar region showing minimal expansion until 230°C. Thermal stress modelling revealed that the GT transition achieved a smoother stress gradient, and no stress concentrations were detected in either transition type under working conditions.
- (3) The GT FGM showed higher strength but scarifying almost half ductility compared to the AT FGM. Both FGMs fractured in the Invar region, with the GT sample exhibiting lower measured elongation due to gauge length effects. Reported ductility values should be considered indicative rather than absolute.

This study provides a more cost-effective design approach, allowing the transition region design to be simplified in industrial applications, thereby reducing manufacturing costs without compromising performance. This provides a new direction for future FGM design, where process optimisation can reduce the complexity of transition region design while improving production efficiency in practical applications.

CRediT authorship contribution statement

Stewart Williams: Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Jialuo Ding:** Writing – review & editing, Supervision, Project administration, Funding acquisition. **Yao Lu:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Goncalo Pardal:** Writing – review & editing, Supervision, Project administration, Funding acquisition. **Guangyu Chen:** Software, Data curation. **Romali Biswal:** Software, Data curation. **Jun Wang:** Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The authors declared that they have no conflicts of interest and have no commercial or associative interest that represents a conflict of interest in connection with the work submitted.

Acknowledgement

This work is financially supported by the “Enabling Multi-Functional Performance through Multi-Material Additive Manufacturing” project

(No 862617 – MULTI-FUN) funded by European Union's Horizon 2020 research and innovation programme. The authors would also like to thank Steve Pope, Kris Bramley, and Nisar Shah for their technical support.

Data Availability

Data will be made available on request.

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2024-09-25

Elsevier

<https://doi.org/10.1016/j.addma.2024.104594>

Wang J, Biswal R, Chen G, et al., (2024) On the composition gradient of steel/Invar functionally graded material manufactured by wire-based direct energy deposition. Additive Manufacturing, Volume 96, September 2024, Article number 104594