

Oxide accumulation effects on wire + arc layer-by-layer additive manufacture process

Xiangfang Xu*, Jialuo Ding, Supriyo Ganguly, Chenglei Diao, Stewart Williams

Welding Engineering and Laser Processing Centre, Building 46, Cranfield University, Bedford,
MK43 0AL, UK

Abstract: A maraging steel wall structure was built layer-by-layer to study oxide accumulation mechanisms and the influence of oxides on the subsequent deposition. An online arc welding camera was also applied to investigate the wetting and spreading behaviour of the deposition on different surface conditions. Two maraging steel walls were deposited under torch shielding only and torch plus tent shielding conditions respectively to study the effect of oxides on the mechanical properties. Upon deposition a mixture of Fe, Al and Ti oxides formed, floated to the weld pool surface and accumulated layer by layer, deteriorating the surface condition such that it was rough and porous, which adversely affected the stability of arc and the wetting and spreading process of the weld pool in subsequent layers. The accumulation of oxides added to the uncertainty of the layer dimension and worsened the surface finish to reduce the structural integrity. Despite that the majority of the oxides floated to the weld pool surface, oxides (up to a few hundred nanometers in diameter) were found to be dispersed in the additively manufactured structure and might be one of the strengthening sources resulting in a 11% increase in UTS and a 19% decrease in elongation compared to the structure built in the torch plus tent shielding condition.

* Corresponding author.

E-mail address: xiangfang.xu@cranfield.ac.uk (X. Xu).

Keywords: Wire + arc additive manufacture; Oxides accumulation; Wetting and spreading; Maraging steel

1. Introduction

Herzog et al. (2016) explained that additive manufacturing (AM) was initially developed for rapid prototyping to allow greater design freedom, less material wastage, and shorter lead-time in a layer-by-layer manner. Extensive research was devoted to plastic parts initially and as Atzeni and Salmi (2012) summarised, AM technology makes the economic production of small quantities of final plastic parts more feasible. After decades of development, the research interest has now moved to metals, which is attractive to many industrial sectors such as aerospace, shipbuilding and oil and gas where massive large-scale metallic structures are fabricated.

Almeida and Williams (2010) developed the wire + arc additive manufacture (WAAM) process, aimed at producing massive (meter scale) metallic structures with full density, high deposition rate, and low equipment cost. Ding et al. (2015) described that in WAAM the commercial welding power source is utilised to generate the electric arc as the heat source and an external wire is fed as the feedstock; usually, a preprogrammed robot arm is applied to control the deposition path to build a functional part in a layer-by-layer manner. So far, WAAM has been proved feasible for a broad range of materials from steel, titanium, Inconel superalloy, high strength aluminium to refractory metals, as stated by Williams et al. (2015).

Ever since WAAM has been proposed, various researches have been carried out to fully understand this process. Cong et al. (2014) investigated the effects of power source mode on the porosity characteristic of additively manufactured high strength aluminium alloy with cold metal transfer (CMT) process and studied the relationship between deposition mode and WAAM materials properties (Cong et al. (2017)). The interpass rolling technology has been

applied by Gu et al. (2016) and Colegrove et al. (2013) to WAAM for aluminium and steel to induce recrystallization and eliminate porosities in aluminium and to reduce distortion and residual stress in steel, respectively. Various in-situ inspection of metal AM processes has been applied as well. For example, Berumen et al. (2010) utilized an in-line camera based set-up to measure the dimensions of the melt pool and the mean radiation emitted; Furumoto et al. (2012) assembled a high-speed camera to monitor the consolidation of the metal powder during irradiation. Similar technologies are also being investigated in WAAM now.

In WAAM, two aspects are of most concern: surface finish and mechanical properties, which represents the surface and interior qualities, respectively. A component built by WAAM is built up on a layer by layer basis. Each layer except the first, is formed by solidifying the liquid weld pool on the surface of the previously solidified layers. Therefore, the surface condition and the wetting and spreading behaviour of the weld pool play a dominant role in controlling the surface finish of WAAM parts. The commercial wire used for WAAM is designed for welding with deoxidizing alloying elements (Si, Mn, Al, Ti, etc.) added deliberately to prevent porosity and to minimise the oxygen content remaining in the solidified bead, as described by D.J.Widgery (1976). In conventional welding, these elements combine with oxygen and float to the surface to form scales or slags covering and protecting the inner bead; such scales or slags could be easily removed during multi-layer and multi-pass welding and will not affect the final weld quality.

As reported by Cáceres and Selling (1996), defects such as inclusions, entrapped dross and gas porosity could lead to a decrease in tensile ductility and strength which can be correlated with the area fraction of defects in the fracture surface. In another AM process, selective laser melting (SLM), experiments are carried out in an inert atmosphere in a sealed chamber. Despite this, Kempen et al. (2011) and Yasa et al. (2010) reported that in their research on 18Ni-300 maraging steel Ti and Al combined oxides (TiO_2 : Al_2O_3) were present as large

inclusions (in the size of 10-20 μm) in the matrix. These inclusions reduced the toughness especially in the aged condition when the material was brittle. Thijs et al. (2011) applied microscopy and spectroscopy to investigate the initiation of these oxides (size ranging from 10-120 μm). It was found that an oxide layer containing Al and Ti was created on top of each layer; upon depositing the next layer, the oxide layer was broken and dragged further. Such accumulation of oxides in the bulk resulted in the large and irregularly shaped inclusions inside the final component.

Similarly to SLM, WAAM is also a layer-by-layer process so oxide accumulation could be an issue, the only difference being that the main source of oxide in the SLM process is the powder (oxide surface, humidity, entrapped gas and so on) while in the WAAM process the atmosphere is the most significant source of oxygen. Thus, oxides formation mechanism and how oxides will affect the surface finish and mechanical properties of WAAM parts have been studied in this research. Furthermore, WAAM is characterised by a much larger weld pool and a much slower cooling rate compared to SLM. Therefore for WAAM wetting and spreading of the liquid weld pool on the as-solidified surface will be a significant issue determining the surface finish. Furthermore, the slow cooling rate allows enough time for the low-density oxides to escape from the inner weld pool and float to the surface. Successive thermal cycles lead to the part being built being kept at a high temperature for long periods, increasing the likelihood of forming more undesirable oxides on the surface.

A common practice in AM is to melt the metallic powder or wire in an inert atmosphere; however few researches have been carried out to investigate the benefits of this. Customising an inert atmosphere and reducing the oxygen level to a certain low level is not always practical and economic and could be time-consuming especially when manufacturing large-scale parts. The preceding inclusions issue found in SLM showed that it is hard to eliminate

oxides through the use of a low oxygen atmosphere so more understanding needs to be gained on how these oxides could affect the AM process.

AM research has been very focused on achieving mechanical properties comparable to the wrought materials, with relatively few studies specifically on understanding the influence of surface conditions on subsequent layer deposition and the resultant surface finish and mechanical properties of the components. Therefore, this paper aims to investigate the formation and accumulation mechanisms of oxides and effect of oxides on the wetting and spreading of the deposition, arc stability and structure integrity and mechanical properties of the WAAM products.

2. Materials and methodology

The WAAM system comprises a welding power source, a wire feeding system, a tent (filled with pure Ar when required, as shown in Fig. 1b), a 6-axis ABB robot with a welding torch mounted, and a working platform. A 10 mm thick mild steel plate was used as the substrate, and the 1.2 mm maraging steel wire (MARVAL 18S, equivalent to maraging 250-grade steel) was used as the feedstock. The chemical composition of the wire is listed in Table 1.

Table 1 Chemical composition of the MARVAL 18S filler wire (wt. %).

Ni	Mo	Co	Ti	Al	C	Si	Mn	Fe
18.28	4.69	8.21	0.44	0.11	<0.01	<0.1	<0.1	Balance

The experimental procedure consists of two phases to represent two different WAAM processes. In phase 1, a plasma power source and a plasma torch were assembled to the robot arm to apply the plasma-WAAM process in an open atmosphere (Ar for plasma gas: 0.8 l/min, Ar for shielding gas: 10 l/min). A 9-layer wall structure was built in the form of stairs (see Fig. 1a) in which the length of each layer was reduced by a pre-determined value (10 mm) to investigate oxides accumulation process. An online arc welding camera (Redman MC500) was attached to the torch to investigate the formation of oxides and monitor the wetting and

spreading behaviour of the deposition on different layers (as shown in Fig. 1a). Meanwhile, an AMV 5000 arc monitor was deployed to monitor the arc voltage variation pattern in real time. In phase 2, a CMT power source and a CMT torch were fixed to the set-up to perform the CMT-WAAM process. Two 6-layer walls were built in comparison: one in the open atmosphere with standard torch shielding only and the other one in an argon-filled tent (where oxygen level was controlled below 300 ppm by using Z230 Oxygen Analyser, Hitech Instruments and the measurement was taken at the bottom of the tent) to investigate the influence of the ambient condition on the surface finish and structure integrity of the WAAM structures, as shown in Fig. 1b. Two additional walls were built under identical conditions and tensile samples were sectioned across the layers to study the effect of oxides on the mechanical properties of the WAAM products. Three samples in each wall were tested for averaging.

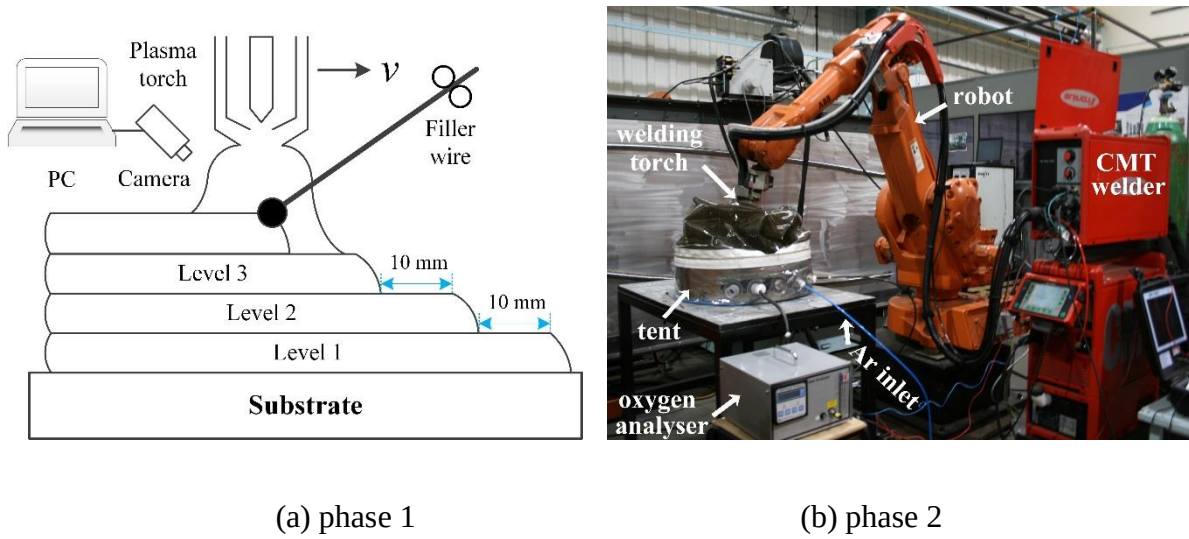


Fig. 1. Experimental setups of the WAAM process.

Samples were cross-sectioned in a plane perpendicular to the substrate and the wall length direction to examine different layers of deposit and a procedure consisting of mounting, grinding and polishing were followed to prepare the specimens for oxide analysis. Scanning Electron Microscope (SEM, FEI XL30-SFEG) equipped with the energy-dispersive

spectrometry detector (EDS, Oxford Instrument, acquisition time 60s, acceleration voltage 10kV) was used to analyse the morphology, thickness and chemical composition of the oxide layers on top of the different layers of the wall structure. The Zwick/Roell hardness tester was utilised to measure the microhardness under a load of 1 kg and holding time of 15s. Tensile coupons were designed and machined as per ASTM-E8 standard, and the tensile test was carried out with Instron 5500R electromechanical testing machine (crosshead speed: 0.5 mm/min) at room temperature.

3. Results and discussion

3.1 Morphology of the oxide layer

Fig. 2 shows the macro-morphology of the oxide layers on the top of different levels in the open atmosphere. Each level of the stair-shaped wall consists of a different number of deposition layers ranging from 1 to 9. The surface of the first layer is very smooth though the dark colour of the bead reveals the surface oxidation. As the number of layer increases, the top surface progressively becomes rougher with different orientation of the oxide scales with different colours (9 layers).

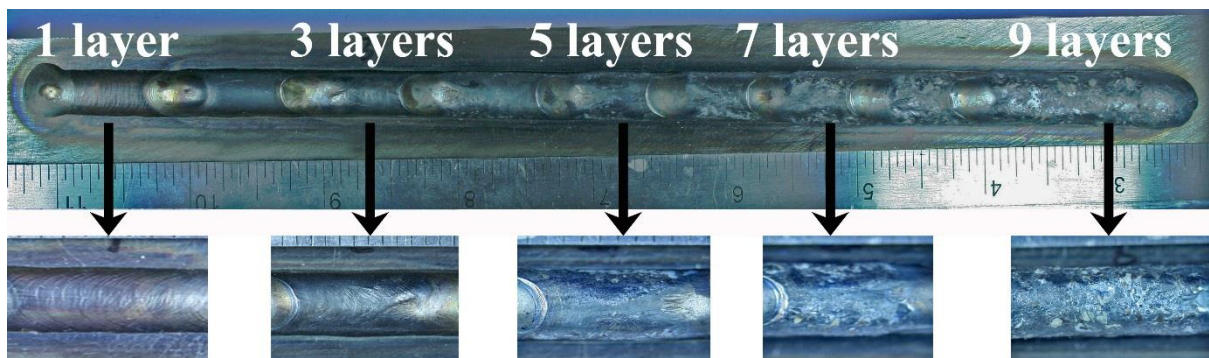
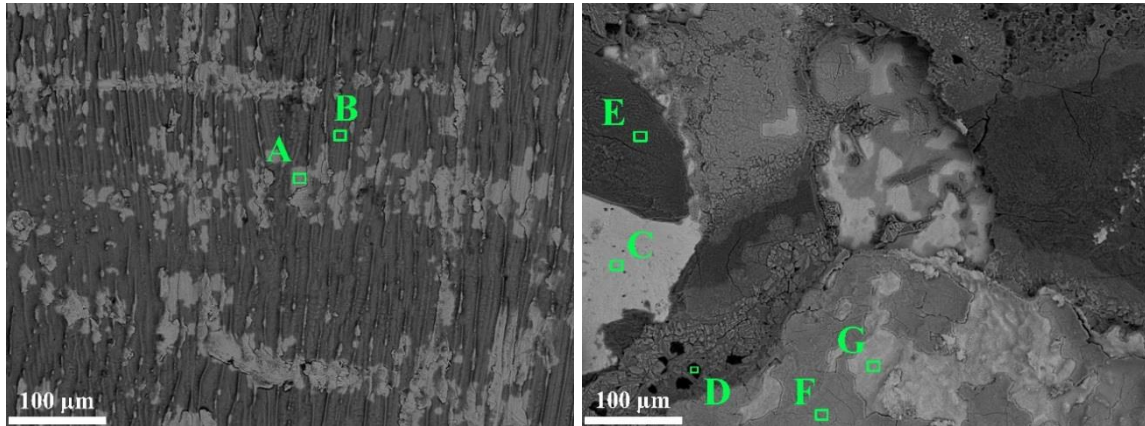


Fig. 2. Macro-morphology of oxides (phase 1, wire feed speed (WFS) = 1.8m/min, travel speed (TS) = 0.2m/min).

Fig. 3 presents the micro-morphology of the oxide layer at different levels from the top view through SEM (in secondary electron mode, SE mode). As can be seen, the oxides

morphology and constituents at the early stage (1 layer) are rather simple with only two kinds of distinct regions (Fig. 3a): the dark regions are the oxidised areas while the bright regions reveal the exposed metal. In comparison, the surface of the 9th layer deposit contains complex oxides which are revealed by the contours of different contrasting regions under SEM (Fig. 3b).



(a) 1 layer

(b) 9 layers

Fig. 3. Micro-morphology of oxides (Acc.V=10kV, Spot=3.0, Magn=200, Det=SE, WD=4.8).

Table 2 shows the EDS analysis results of different regions as labelled in Fig. 3. It was found that the whiter area is less oxidised compared to the darker area and as the number of layers increased, more areas containing different oxygen levels (ranging from 9.97% to 47.43%) and various combinations of oxides were found non-uniformly distributed throughout the oxide layer. Given the elemental content of the filler wire, the oxides were primarily comprised of Al oxides, Ti oxides, and Fe oxides. It should also be noted that among oxide islands there are also some areas where the oxide scale is dislodged, resulting in the exposure of fresh metallic surface e.g. Zone C in Fig. 3b. Such areas, on the one hand, separate oxides islands and leads to increased roughness and thickness variation; on the other hand, would have a lower melting point compared to oxides, which may result in non-uniform melting-

floating phenomenon during the transition of the weld pool. This will be further explained later.

Table 2 EDS analysis of marked areas from Fig. 3 (wt. %).

	Spectrum Label	O	Al	Ti	Fe	Co	Ni	Mo
1 layer	A	2.12	-	0.28	70.7	6.07	14.64	3.23
	B	16.21	-	0.63	63.36	5.46	11.13	3.19
9 layers	C	-	-	0.94	66.11	8.96	18.85	5.14
	D	43.98	3.63	25.67	25.89	-	-	-
	E	47.43	10.38	36.28	4.96	-	-	-
	F	24.89	-	-	62.41	6.68	5.33	0.69
	G	9.97	0.21	0.27	40.66	9.71	32.08	7.09

3.2 Oxide accumulation mechanism

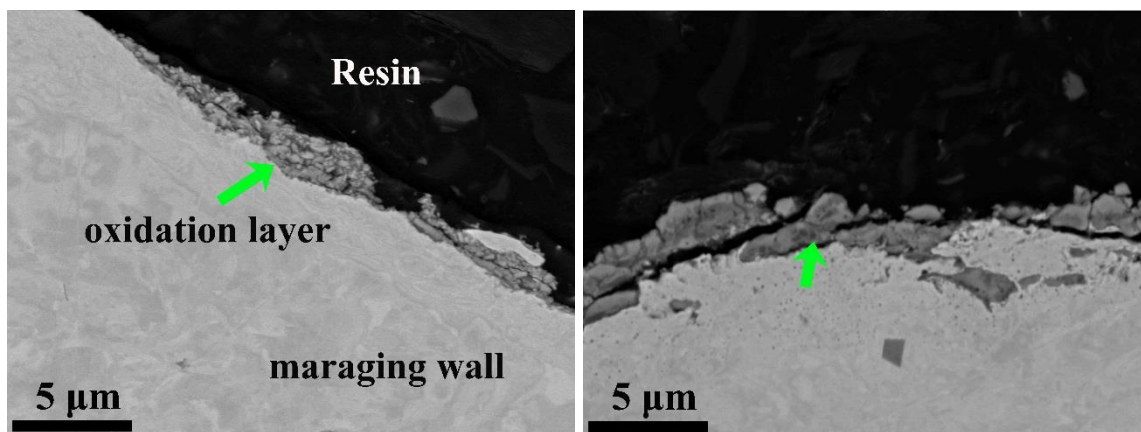
Fig. 4 shows the SEM images of the cross-section of each level, which clear displays the oxides accumulation process. When there is only one layer, a few oxides form a thin oxide layer and do not cover the entire surface (Fig. 4a). As more layers have been deposited, the oxide layer gradually grows and covers the entire surface and thickness increases, varying from 1.3 μm to 13.04 μm when the number of layers varies from 1 to 9 (Fig. 4f).

The oxide layers are rough and porous and do not distribute uniformly on the top of each layer. They are unstable structures with limited adherence to the matrix, and may break with expansion and contraction of the base metal and oxides due to the accumulation of thermal stress. Such stress develops from the mismatch of coefficient of thermal expansion (CTE) between the oxides and the matrix, as explained by Guleryuz and Cimenoglu (2009). Also, oxides change phase with temperature and elemental diffusion and may decompose into several sub-layers as shown in Fig. 4e.

Generally, oxides stay on the surface of the deposits as big scales and oxide ingress is not observed within the deposited material which is clean and free from oxides. However, on

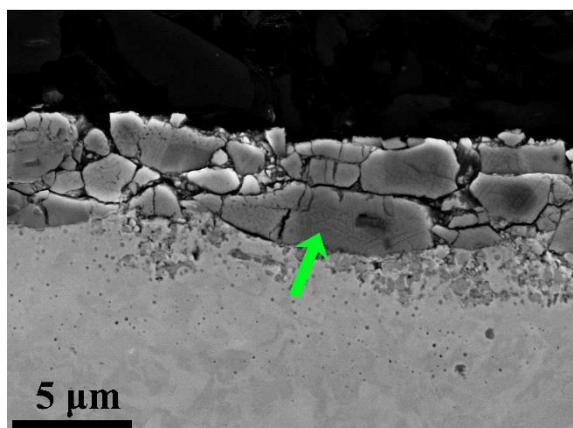
occasions, oxides are found to be trapped as an inclusion during the solidification process as shown by the red arrow in Fig. 4d.

Fig. 4g shows the SEM linear scanning results from the maraging steel wall matrix to the oxide layer (as shown in Fig. 4d). It is clear that there are significant variations of the elemental contents of Fe, Al, Ti and O from the matrix to the oxide layer with the Al, Ti and O content increasing and Fe content decreasing. From the outer surface of the oxide layer to the matrix, the Al, Ti and Fe oxides enrichment are observed in sequence. This could be due to different densities and diffusion directions and diffusion rates of oxides, as indicated by Guleryuz and Cimenoglu (2009) since they are of different thermodynamics and kinetics. In successive layers, the Al and Ti oxides decompose under the high arc temperature and, after recombining with the additional oxygen they float to the surface and accumulate as fresh scales. In contrast, the iron oxides form in the solid state when the diffusion of atoms is minimised and the availability of Fe atoms is much higher beneath the Al and Ti oxides.

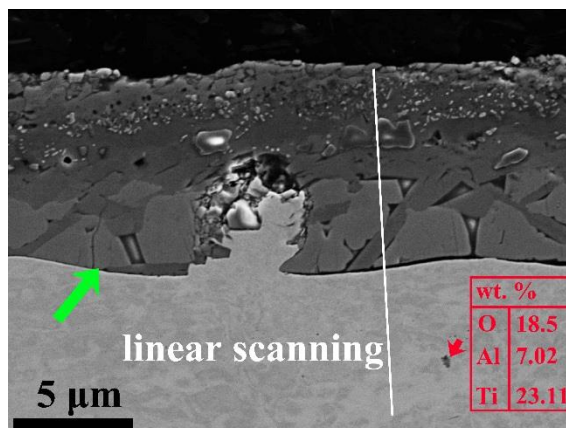


(a) 1 layer

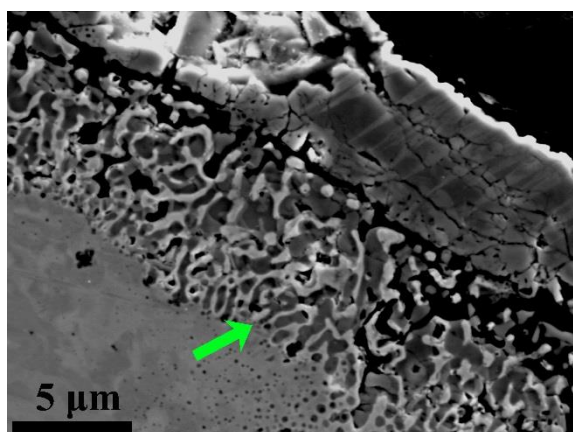
(b) 3 layers



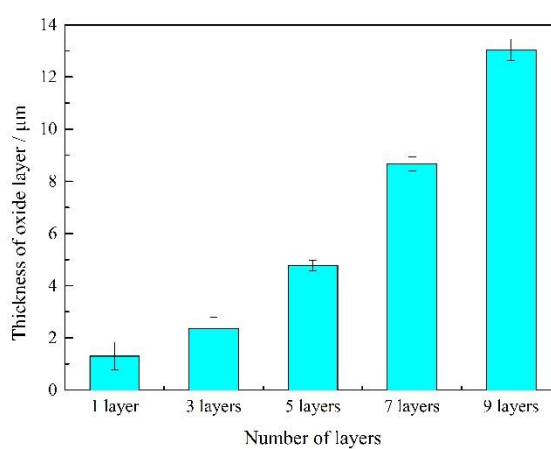
(c) 5 layers



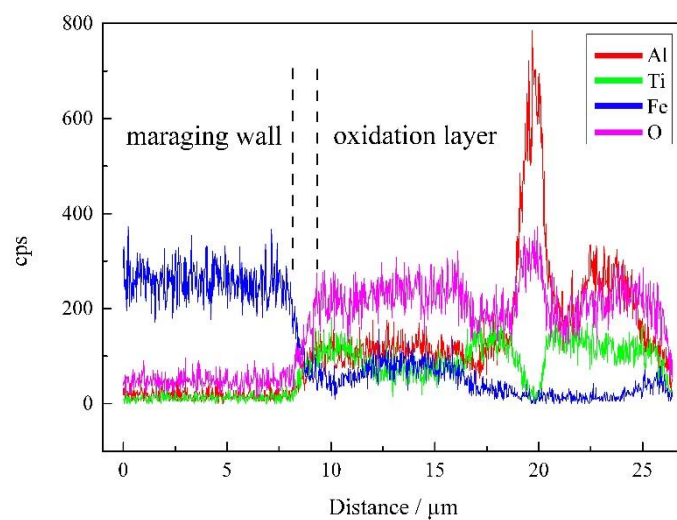
(d) 7 layers



(e) 9 layers



(f) thickness of oxide layer (error bar: standard deviation)



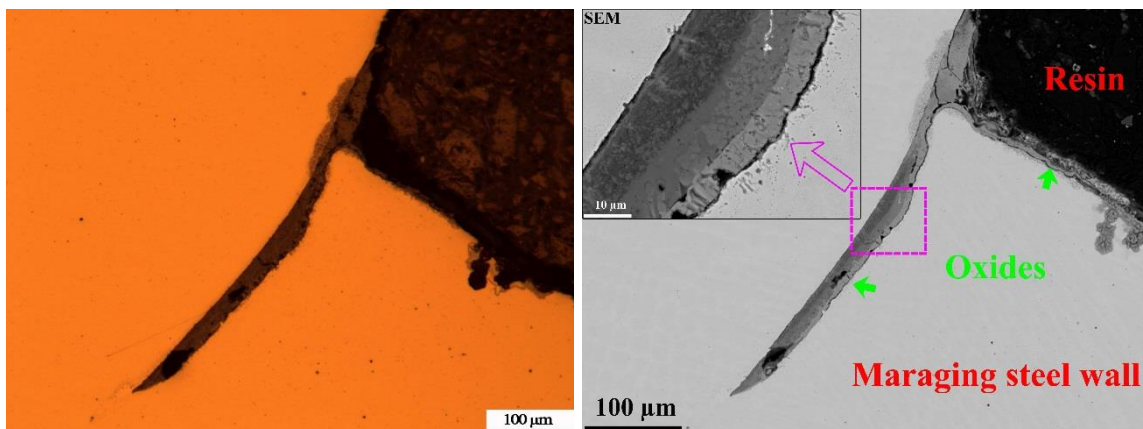
(g) linear scanning results from Fig. 4d

Fig. 4. Cross-sections of oxide layers on each level, thickness of oxide layers and elements distribution (for a-e, Acc.V=10kV, Spot=3.0, Magn=5000, Det=SE, WD=4.8).

3.3 Oxides entrapped between layers

Oxides after formation would float to the weld pool surface due to their lower density compared with the density of the liquid metal pool. However, depending on the liquid flow of the melt pool, oxides could be found to be trapped within the liquid melt pool. This problem is likely to be exacerbated with increasing numbers of layers when the scale thickness increases which may lead to the entrapment of oxides between two successive passes, as shown in Fig. 5a. The oxide zone is embedded at the interface where the two layers join and extends from the surface into the as-deposited structure up to about 200 μm . Although the penetration could be neglected when compared to the surface waviness of the WAAM parts as such oxides will be removed during standard machining, the presence of such unexpected oxides still leads to the structural and elemental discontinuity in the WAAM structure.

Figs. 5b-f show the SEM elemental mapping results of the trapped oxide zone. Similarly to the oxides on the top surface, these inner oxides also have a multilayered structure, the composition of which is complex and not uniform. Different regions rich in Ti, Al and Fe are located in different layers.



(a) OM

(b) SEM

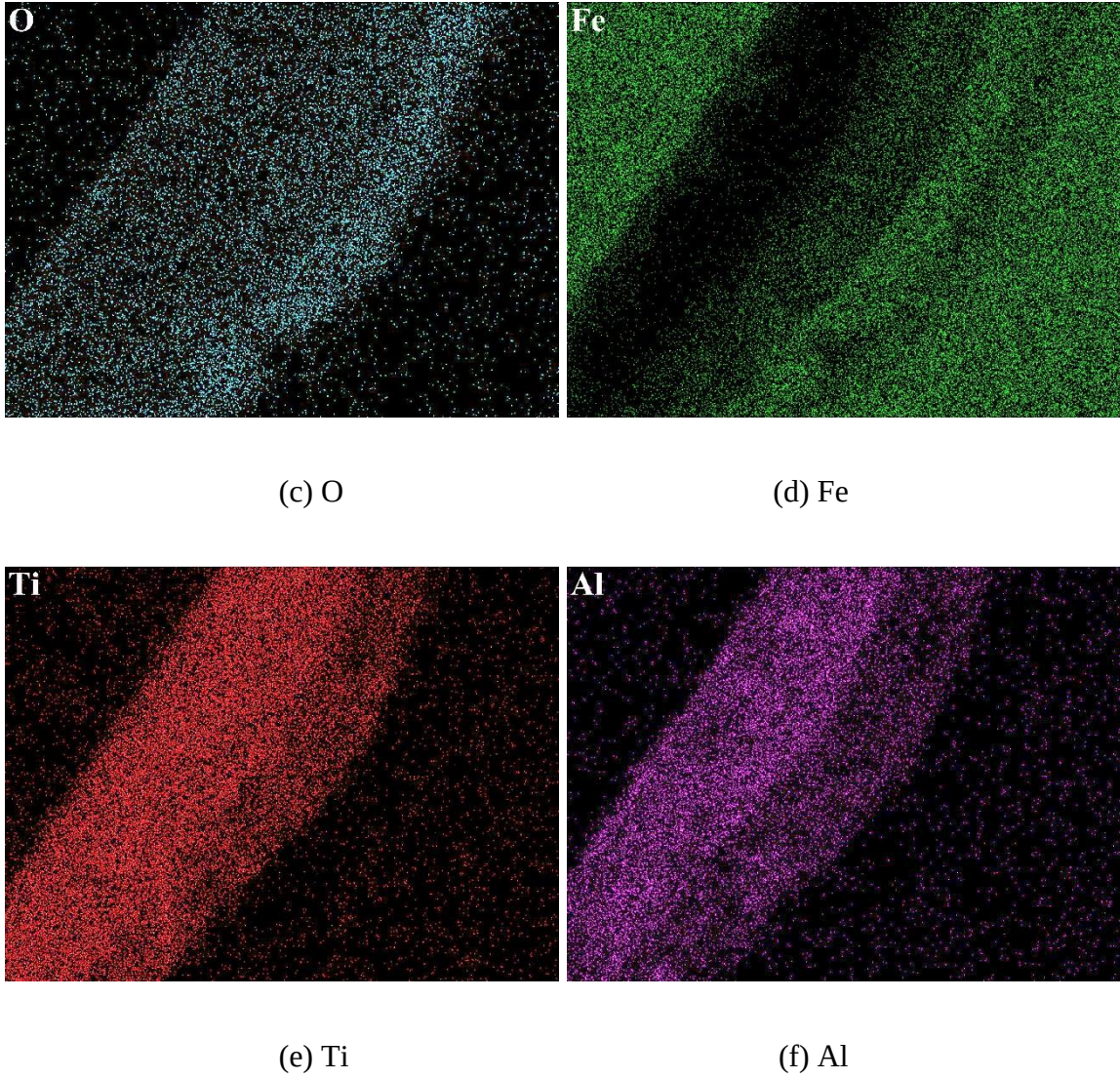


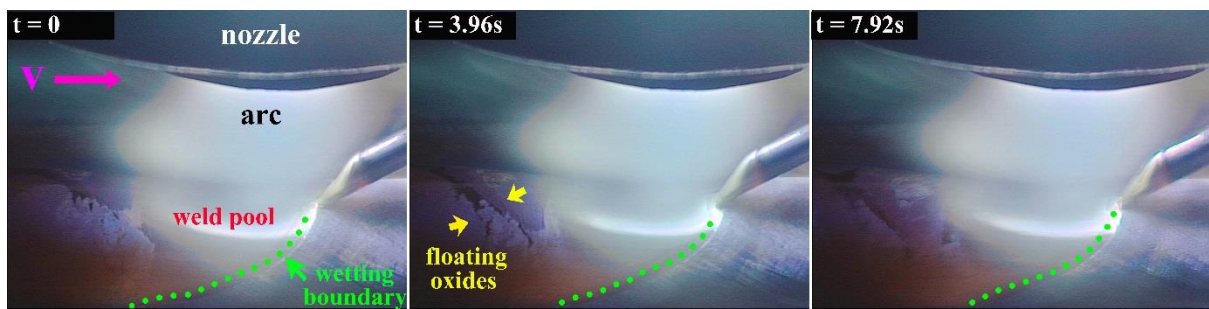
Fig. 5. Entrapped oxide layer found between layer 6 and layer 7 (a,b) and its SEM elemental mapping results (c,d,e,f).

3.4 Wetting and spreading on the oxide layer

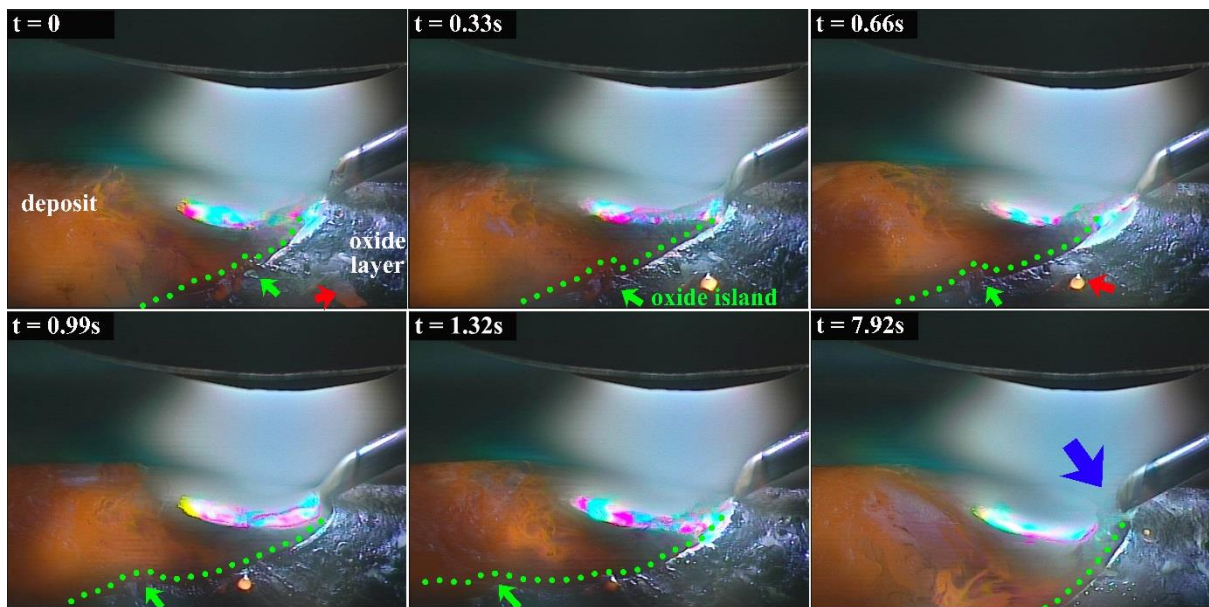
Fig. 6 shows still images from footage of the deposition process on the top of 2nd and 8th layers to represent the wetting and spreading behaviour of the weld pool on a smooth and a rough surface condition, respectively. When depositing on a smoother surface, the curved line indicating wetting boundary maintains a temporally constant profile (indicated by the green dotted line in Fig. 6a), indicating a steady wetting and spreading process of the 3rd layer.

Besides, the relatively smooth curve also implies the constant resistance from the surface against the weld pool along the depositing direction.

In comparison, when depositing on a rough surface, the wetting boundary shows temporal variation and shifting depending on the surface condition at the weld pool boundary: the rougher the surface, the greater variation of the curve (Fig. 6b). This is because the rough surface would alternate the arc length which would change the total thermal energy applied which will impact the weld pool dimension and the wetting angle.



(a) on the 2nd layer (on a relatively smoother surface)



(b) on the 8th layer (on a relatively rougher surface)

Fig. 6. Footage of deposition process on different surface conditions.

Bonn et al. (2009) reported that for the wetting and spreading on a smooth surface (as shown schematically in Fig. 7a), the equilibrium system should conform to the following balance of forces to minimise the free energy of the system,

$$\gamma_{SG} = \gamma_{SL} + \gamma_{LG} \cdot \cos \theta \quad (1)$$

Where γ_{SG} is the solid-gaseous surface tension, γ_{SL} is the solid-liquid surface tension, γ_{LG} is the liquid-gaseous surface tension and the θ is the equilibrium contact angle.

For a particular solid-liquid-gaseous system the three surface tensions can be considered constant. Hence the equilibrium contact angle θ remains unchanged during the deposition on a smooth surface, which leads to a stable wetting and spreading process of the weld pool and the constant wetting boundary as indicated in Fig. 6a.

In contrast, when depositing on the rough surface, the variation of solid surface orientation (as shown in Fig. 7b) leads to the modified equilibrium equation as below,

$$\gamma_{SG} \cdot \cos \alpha = \gamma_{SL} + \gamma_{LG} \cdot \cos \theta' \quad (2)$$

Where α is the angle of the rough surface compared to the smooth surface.

On the rough surface, α is unpredictable, so the wetting angle θ' keeps changing to balance the forces and to establish a new equilibrium system, which is the driving force of the adaptive wetting boundary as shown in Fig. 6b. When α is positive (say, a convex oxide island blocking the way, Fig. 6b) more resistance is applied to the weld pool to prevent the spreading; when α is negative (say, a concave on the surface), the gravity-induced dragging force helps the wetting and spreading process. The wetting and spreading process in WAAM process on the rough surface is such an alternating manner between “dragging” and “blocking” in an adaptive mode over the deposition.

When depositing on such an oxide layer, oxides will float to the surface of the weld pool (as shown in Fig. 6a by yellow arrows) due to their higher melting point and lower density compared to the matrix. However, when facing a large oxide island (as indicated in Fig. 6b), instead of floating oxides, the weld pool bypasses oxide islands and changes the wetting boundary to an irregular curve until the weld pool entirely passes it. Besides, even before the weld pool arrives, the material beneath the thinner oxide layer (as indicated by the red arrow in $t=0$, Fig. 6b) and the less oxidized area among the oxide layer (as indicated by the red arrow in $t=0.66$, Fig. 6b) will melt first, even before oxides float. As a consequence, the weld pool wets and spreads on a rough solid surface with dispersed liquid zones, which gives rise to a higher uncertainty of the position of the wetting boundary and bead shape; defects, such as lack of fusion may result from this.

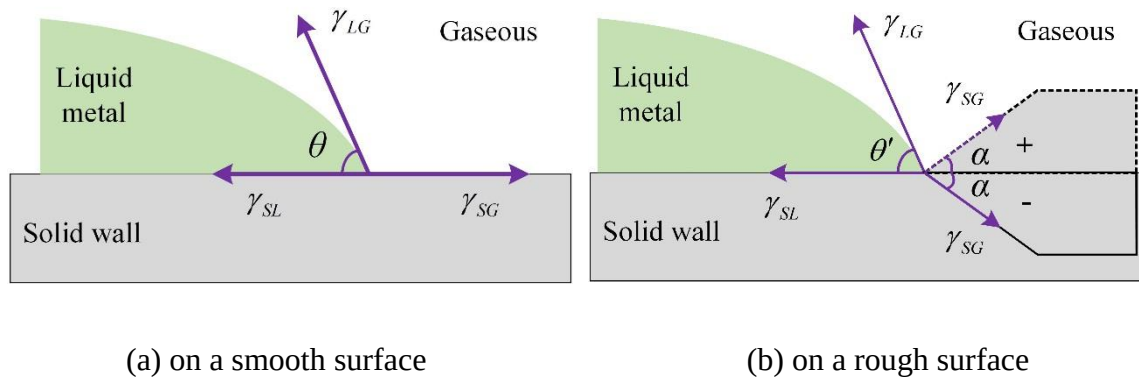


Fig. 7. Wetting and spreading on different surface conditions.

3.5 Arc behaviour on an oxide layer

Miao et al. (2014) reported that the arc behaviour greatly affects the weld pool stability due to the interaction of the different forces between the arc and the weld pool. Furthermore, a consistent metal transfer mode is also essential for the weld pool stability, as concluded by Miao et al. (2015). As with welding, the arc stability is dominant in controlling the deposition shape and quality in WAAM. The accumulated oxides have a significant effect on the arc behaviour. A smooth surface keeps the arc length and the weld pool dimension constant

ensuring a constant wire feeding location with respect to the weld pool over the entire deposition length. It also provides a constant metal transfer mode (ideal liquid bridge mode as shown in Fig. 6a) minimising any disturbance to the weld pool. In contrast, when depositing on the rough surface, the arc length and change of wetting boundary dimension and profile lead to variations in wire feed position with reference to the as-deposited structure. In the plasma-WAAM process, the metal transfer mode depends heavily on the wire feeding position; thus, during the deposition process the liquid bridge can be unstable and could break (as indicated by the blue arrow in Fig. 6b, $t=7.92s$) at some point, or even transfer to globular transfer mode, which introduces more disturbance to the weld pool.

The stability of the arc behaviour is also indicated by the arc voltage. As shown in Fig. 8, the arc voltage of deposition on a smooth surface is much more stable than that of a rough surface with less fluctuation in the waveform. This is because oxides have different electron emission capabilities compared to the matrix and the waviness of the oxidised surface leads to arc length variation and therefore fluctuation of the arc voltage. Considering the constant current (CC) mode in the present WAAM-Plasma process, the heat input is also fluctuating at the same time as the voltage.

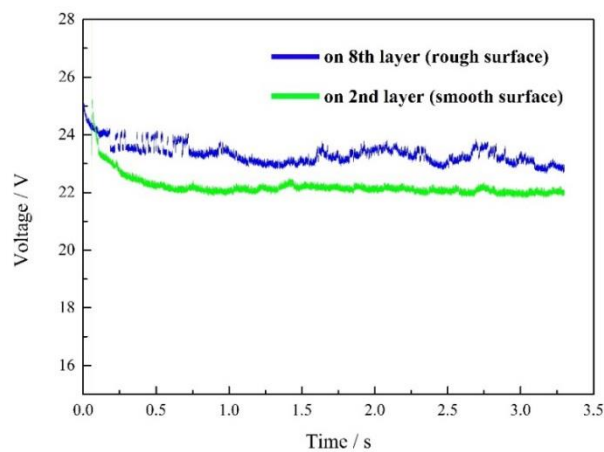
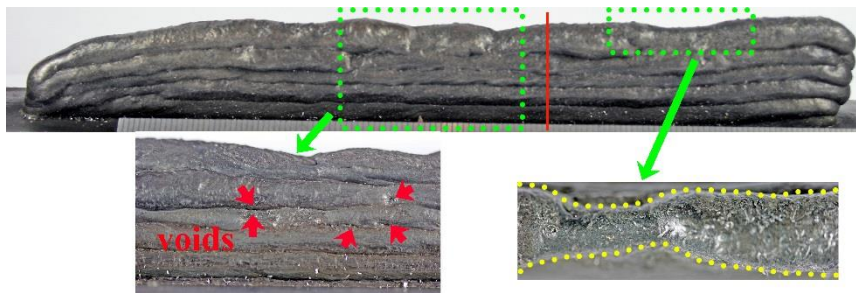


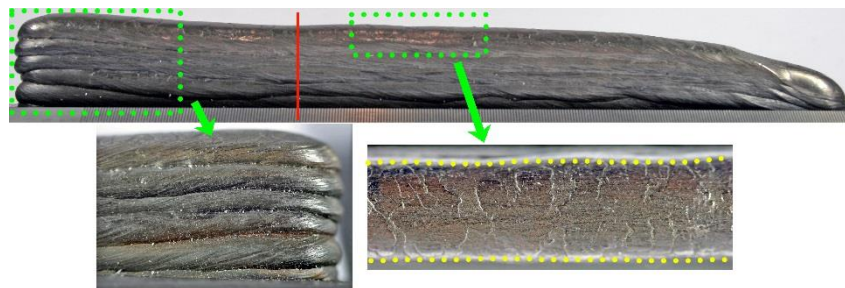
Fig. 8. Arc voltage variation on different surface conditions.

3.6 Structural integrity

The effects of oxides are not only observed in Plasma-WAAM process but also in CMT-WAAM process. Fig. 9 presents two 6-layer walls built by CMT-WAAM process with torch shielding only (open atmosphere) and torch shielding plus inert environment (by depositing within a tent filled with Ar gas) conditions with the same parameters (wire feed speed = 8m/min, travel speed = 6mm/s). In the torch shielding only condition, oxides accumulate layer by layer, leading to a dark and rough surface of the wall structure; irregular bead shape, uneven layer surface, high waviness and voids in and between layers are observed (Fig. 9a). In contrast, when sufficient shielding is guaranteed, the wall structure is shiny with an almost equal width along the length direction; besides, the layer surface is flat with only a few oxides islands, and the waviness is reduced significantly (Fig. 9b). This is because the extra shielding prevents the as-solidified material from being exposed excessively to oxygen at high temperatures, thereby reducing the amount of oxides and providing a better surface condition for successive depositions.



(a) torch shielding only



(b) torch + tent shielding

Fig. 9. Bead shape of WAAM walls in different shielding conditions (phase 2, WFS=8m/min,
TS=0.42m/min)

The transverse cross-sections of the two preceding walls are displayed in Fig. 10 (section positions were indicated by the red line in Fig. 9). The wall with torch shielding only (Fig. 10a) is thinner and the edge is much more irregular compared to the torch plus tent shielding (Fig. 10b).

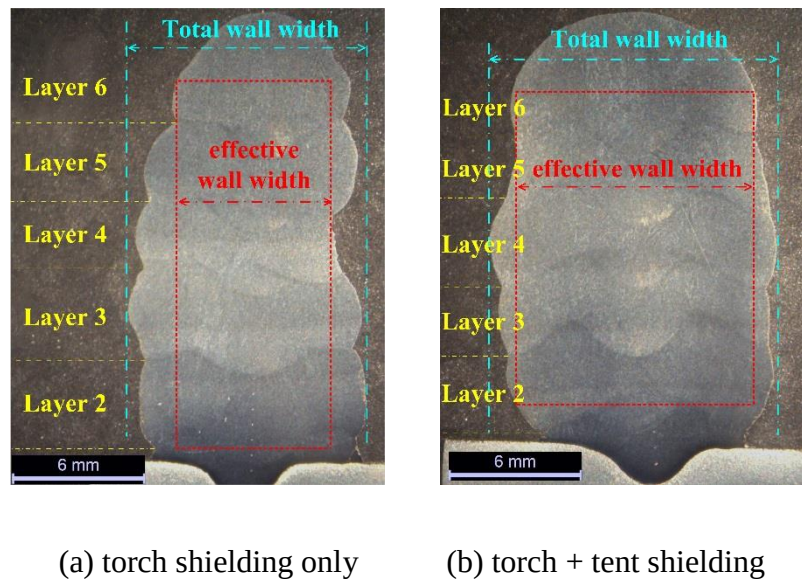


Fig. 10. Transverse cross-section of the two walls (from Fig. 9).

A detailed measurement of the wall dimensions is presented in Table 3. It can be seen that the surface waviness (as defined by Martina et al. (2012)) of the torch-shielding wall is much greater than the counterpart (1.9mm versus 1.2mm). In consequence, extra tent shielding helps to improve the surface finish and the deposition efficiency (from 64.6% to 70.7%). When more layers are deposited, such an improvement would be even more significant.

Table 3 Measurements of the two walls.

		torch shielding only	torch + tent shielding
width	total wall width (TWW) / mm	10.8	13.0
	effective wall width (EWW) / mm	7.0	10.7
	surface waviness (SW) / mm $SW = (TWW - EWW) / 2$	1.9	1.2

area	total wall area (TWA) / mm ²	179.0	214.3
	effective wall area (EWA) / mm ²	115.7	151.5
	deposition efficiency (DE) / % $DE = EWA/TWA$	64.6	70.7

3.7 Structural quality

3.7.1 Microstructure

Even though the surfaces of the two walls are significantly different due to different levels of oxidation, the bulk materials are more or less the same: a combination of cellular and dendritic martensitic microstructure with the dendrites growing perpendicularly to the fusion boundary, as shown in Fig. 11. Besides, no visible oxides in the form of inclusions are found in the cross-sections.

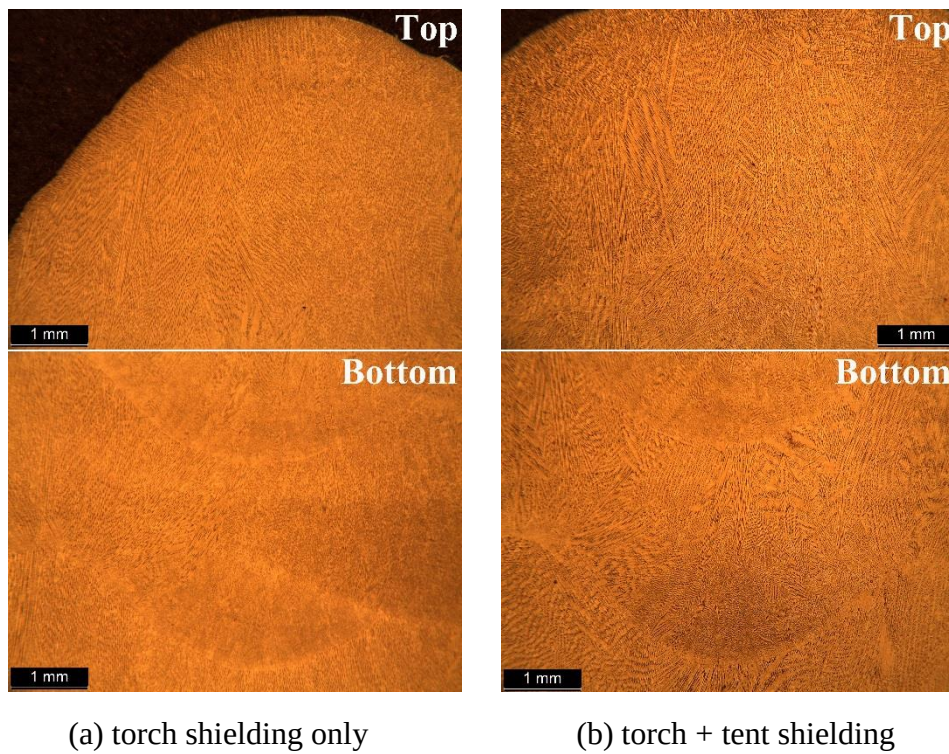


Fig. 11. Microstructure of the two walls.

3.7.2 Microhardness

The microhardness measurements of the two walls in both as deposited and aged (3 hrs at 482°C) conditions along the central line from the top to the bottom are presented in Fig. 12.

In the as deposited condition, both walls show a noticeable hardness increase from the top to the bottom (from around 33 HRC to around 43 HRC) due to the aging effect from successive depositions; after aging, the hardness variation is eliminated and the hardness of both walls is increased to around 52 HRC (52.5 HRC with extra tent shielding, 52.1 HRC without) which is comparable to the aged wrought maraging steel of the identical grade (50-55 HRC, Arai (1991)). Despite the slight variation in values, the effect of oxide accumulation on the microhardness is negligible since the bulk materials of the two six-layer walls underwent identical thermal cycles and therefore resulted in very similar microstructural characteristics.

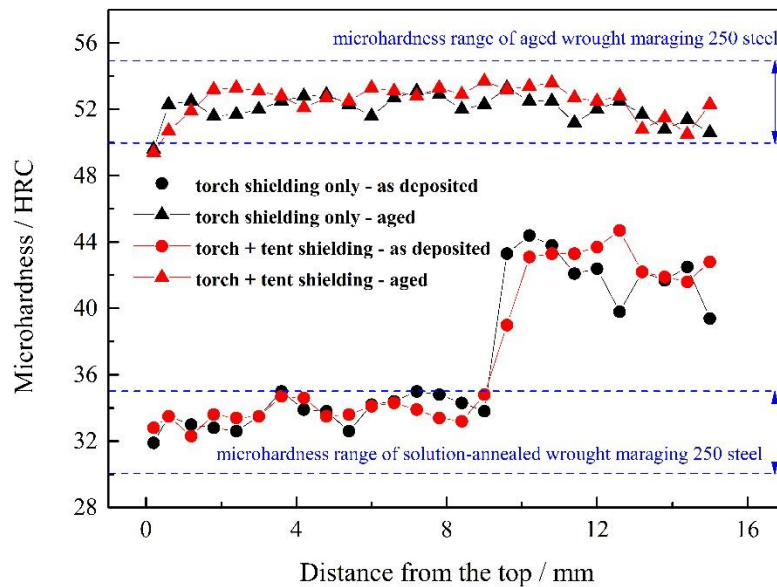


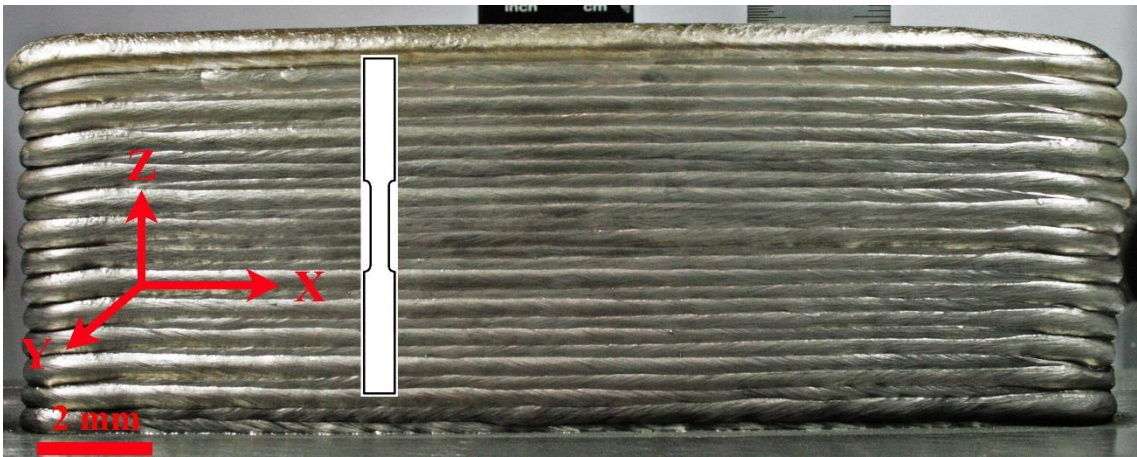
Fig. 12. Microhardness of the two walls from the bottom to the top.

3.7.3 Tensile test

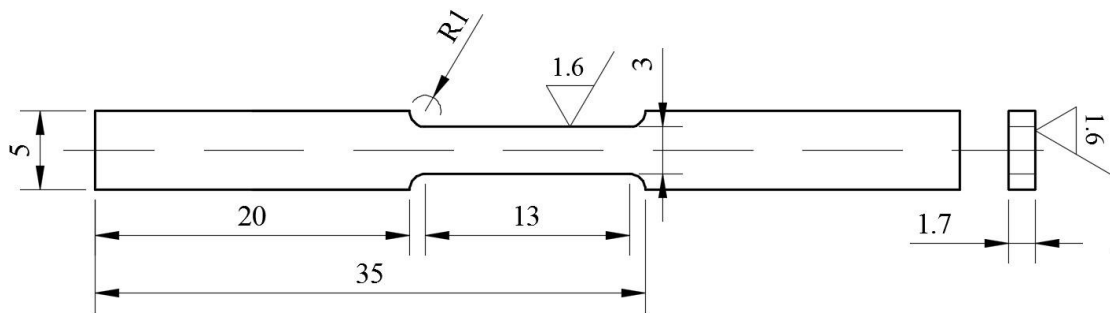
Fig.13 presents the two additional walls built for the tensile test with CMT-WAAM process. The wall built with torch shielding only displays great waviness on the surface (Fig. 13a), and during deposition, especially after a certain number of layers the weld pool collapses from time to time as the accumulated oxides worsens the surface condition. In comparison, the wall built with torch plus tent shielding shows a much better surface finish with regular layer geometries and an almost constant layer height increment.



(a) torch shielding only (wall size: 180×68×9.8mm)



(b) torch + tent shielding (wall size: 185×68×11.3mm)



(c) tensile test specimen drawing

Fig. 13. Two additional walls built for the tensile test and the tensile specimen drawing

Tensile samples were taken along the Z direction, as shown in Fig.13b. Fig. 14 presents the tensile test results. It can be seen that the UTS value of the material deposited with torch shielding only is 11% higher (1165 versus 1048 MPa) than the torch plus tent shielding condition, while the elongation is 19% lower (13.7% versus 17.0%) than the latter.

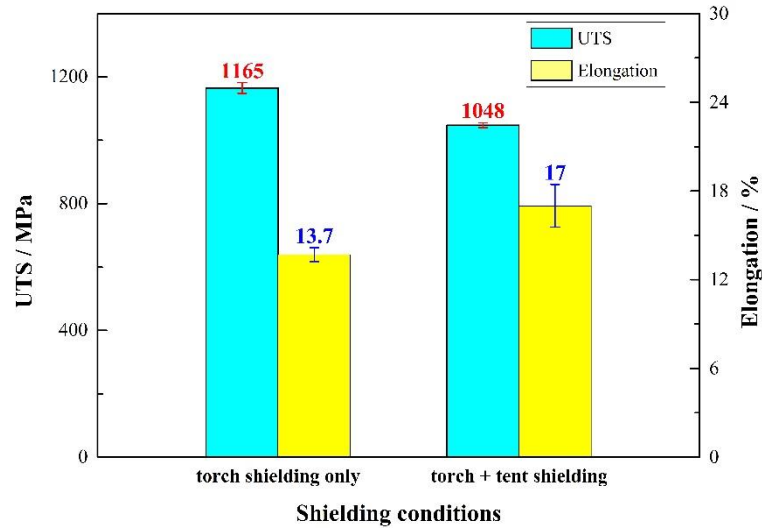
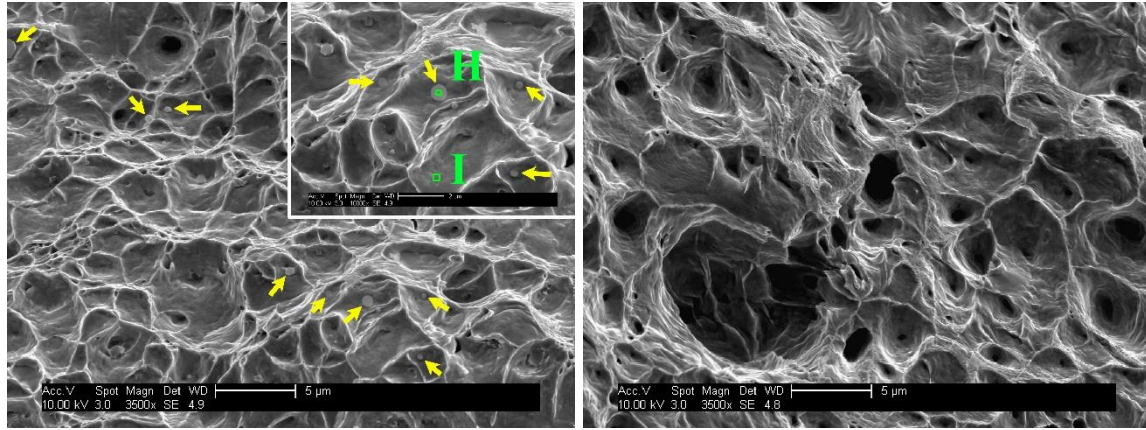


Fig. 14. Tensile test results (error bar: standard deviation)

3.7.4 Fracture surface

Fig. 15 shows the fracture surface morphology of two typical tensile samples from the preceding two walls. A few dispersed spherical particles (a few to a few hundred nanometres in size) are found in the fracture surface of the material deposited with torch shielding only, whereas they are not observed in the torch plus tent shielding condition. Further EDS analysis proves that those particles are mainly Al, Ti and Fe oxides, as shown in Table 4.



(a) torch shielding only

(b) torch + tent shielding

Fig. 15. Fracture surface of tensile specimens.

Table 4 EDS analysis of marked areas from Fig. 15a (wt. %).

Spectrum Label	O	Al	Ti	Fe	Co	Ni	Mo
H	42.5	39.42	4.76	11.19	0.15	-	1.97
I	-	0.81	-	69.03	8.68	17.85	3.63

3.7.5 Oxide strengthening mechanism

The strength difference between the two walls could be due to a number of factors, such as grain size, precipitate size and amount, and fraction of retained austenite. In the present study, the two walls were built with identical parameters except for the shielding condition, therefore oxides could be one of the strengthening sources. Oxides trapped in the material can have different effects which primarily depends on their size and distribution. When trapped in the form of large scales, oxides will reduce ductility and tensile strength both and will lead to a catastrophic failure very quickly. However, the dispersed oxides (up to a few hundred nanometers in diameter) may result in reinforcement of the matrix material. In a research conducted by Hassan and Gupta (2005), it was found that the microhardness, UTS and ductility of the pure magnesium were increased significantly by adding 1.1 volume percentage of 50 nm sized Al_2O_3 due to the grain refinement induced by the nanosized additions. In the present research, the possible strengthening effect from the oxides (up to a

few hundred nanometers in diameter) could be attributed to the load bearing effect, i.e., the strengthening due to obstacle posed by closely spaced hard particles to the dislocation motion and matrix transferring the load to the nanosized reinforcements, as explained by S.Jayalakshmi and M.Gupta (2015).

4. Conclusions

1. Deoxidizing elements in the wire combine with oxygen to form oxides on the surface of the layer; upon deposition, these oxides decompose, re-form, float to the weld pool surface and accumulate layer by layer with newly formed oxides.
2. Oxides in maraging steel are mainly Al, Ti, and Fe oxides and they distribute in different layers inside the oxide layer depending on their densities and diffusion rates.
3. The oxide accumulation leads to increased surface waviness and adversely affects the wetting and spreading of the subsequent deposition by providing various wetting angles and resistance against the liquid metal.
4. The surface oxides destabilize the arc and weld pool and thereby, the surface finish of the WAAM structure. Extra tent shielding substantially improved the surface waviness and deposition efficiency by 37% and 9%, respectively.
5. Dispersed oxides (up to a few hundred nanometers in diameter) are found to be trapped in the material deposited with torch shielding only and they may be one of the strengthening sources to result in a 11% increase in UTS and 19% decrease in elongation due to the load bearing effect.

Acknowledgement

The authors wish to acknowledge the financial support from China Scholarship Council (CSC) and the WAAMMat program industrial partners. The authors would also like to thank Dr. Xianwei Liu for the assistance during the SEM analysis.

References

- Almeida, P., Williams, S., 2010. Innovative process model of Ti-6Al-4V additive layer manufacturing using cold metal transfer (CMT). *Solid Free. Fabr. Symp.* 25–36
- Arai, T., 1991. Volume 4: Heat Treating, in: *ASM Handbook*. ASM International, p. 2173. doi:10.1016/S0026-0576(03)90166-8
- Atzeni, E., Salmi, A., 2012. Economics of additive manufacturing for end-usable metal parts. *Int. J. Adv. Manuf. Technol.* 62, 1147–1155. doi:10.1007/s00170-011-3878-1
- Berumen, S., Bechmann, F., Lindner, S., Kruth, J.-P., Craeghs, T., 2010. Quality control of laser- and powder bed-based Additive Manufacturing (AM) technologies. *Phys. Procedia* 5, 617–622. doi:10.1016/j.phpro.2010.08.089
- Bonn, D., Eggers, J., Indekeu, J., Meunier, J., 2009. Wetting and spreading. *Rev. Mod. Phys.* 81, 739–805. doi:10.1103/RevModPhys.81.739
- Cáceres, C.H., Selling, B.I., 1996. Casting defects and the tensile properties of an Al-Si-Mg alloy. *Mater. Sci. Eng. A* 220, 109–116. doi:10.1016/S0921-5093(96)10433-0
- Colegrove, P. a., Coules, H.E., Fairman, J., Martina, F., Kashoob, T., Mamash, H., Cozzolino, L.D., 2013. Microstructure and residual stress improvement in wire and arc additively manufactured parts through high-pressure rolling. *J. Mater. Process. Technol.* 213, 1782–1791. doi:10.1016/j.jmatprotec.2013.04.012
- Cong, B., Ding, J., Williams, S., 2014. Effect of arc mode in cold metal transfer process on porosity of additively manufactured Al-6.3%Cu alloy. *Int. J. Adv. Manuf. Technol.* 76, 1593–1606. doi:10.1007/s00170-014-6346-x
- Cong, B., Qi, Z., Qi, B., Sun, H., Zhao, G., Ding, J., 2017. A Comparative Study of Additively Manufactured Thin Wall and Block Structure with Al-6.3%Cu Alloy Using Cold Metal Transfer Process. *Appl. Sci.* 7, 275. doi:10.3390/app7030275
- D.J.Widgery, 1976. Deoxidation Practice for Mild steel Weld metal. *Weld. Res*
- Ding, J., Colegrove, P., Martina, F., Williams, S., Wiktorowicz, R., Palt, M.R., 2015. Development of a laminar flow local shielding device for wire+arc additive manufacture. *J. Mater. Process. Technol.* 226, 99–105. doi:10.1016/j.jmatprotec.2015.07.005
- Furumoto, T., Alkahari, M.R., Ueda, T., Aziz, M.S.A., Hosokawa, A., 2012. Monitoring of Laser Consolidation Process of Metal Powder with High Speed Video Camera. *Phys. Procedia* 39, 760–766. doi:10.1016/j.phpro.2012.10.098
- Gu, J., Ding, J., Williams, S.W., Gu, H., Ma, P., Zhai, Y., 2016. The effect of inter-layer cold working and post-deposition heat treatment on porosity in additively manufactured aluminum alloys. *J. Mater. Process. Technol.* 230, 26–34. doi:10.1016/j.jmatprotec.2015.11.006
- Guleryuz, H., Cimenoglu, H., 2009. Oxidation of Ti-6Al-4V alloy. *J. Alloys Compd.* 472, 241–246. doi:10.1016/j.jallcom.2008.04.024
- Hassan, S.F., Gupta, M., 2005. Enhancing physical and mechanical properties of Mg using nanosized Al₂O₃ particulates as reinforcement. *Metall. Mater. Trans. A* 36, 2253–2258. doi:10.1007/s11661-005-0344-4

- Herzog, D., Seyda, V., Wycisk, E., Emmelmann, C., 2016. Additive manufacturing of metals. *Acta Mater.* 117, 371–392. doi:10.1016/j.actamat.2016.07.019
- Kempen, K., Yasa, E., Thijs, L., Kruth, J.P., Van Humbeeck, J., 2011. Microstructure and mechanical properties of selective laser melted 18Ni-300 steel. *Phys. Procedia* 12, 255–263. doi:10.1016/j.phpro.2011.03.033
- Martina, F., Mehnen, J., Williams, S.W., Colegrove, P., Wang, F., 2012. Investigation of the benefits of plasma deposition for the additive layer manufacture of Ti-6Al-4V. *J. Mater. Process. Technol.* 212, 1377–1386. doi:10.1016/j.jmatprotec.2012.02.002
- Miao, Y., Xu, X., Wu, B., Han, D., Zeng, Y., Wang, T., 2015. Effects of bypass current on arc characteristics and metal transfer behaviour during MIG-TIG double sided arc welding. *J. Mater. Process. Technol.* 224, 40–48. doi:10.1016/j.jmatprotec.2015.04.026
- Miao, Y., Xu, X., Wu, B., Li, X., Han, D., 2014. Effects of bypass current on the stability of weld pool during double sided arc welding. *J. Mater. Process. Technol.* 214, 1590–1596. doi:10.1016/j.jmatprotec.2014.02.029
- S.Jayalakshmi, M.Gupta, 2015. Metallic Amorphous Alloy Reinforcement in Light Metal Matrices. doi:10.1007/978-3-319-15016-1
- Thijs, L., Van Humbeeck, J., Kempen, K., Yasa, E., Kruth, J.-P., 2011. Investigation on the inclusions in maraging steel produced by Selective Laser Melting, in: 5th International Conference on Advanced Research in Virtual and Rapid Prototyping. pp. 297–304. doi:doi:10.1201/b11341-48r10.1201/b11341-48
- Williams, S.W., Martina, F., Addison, A.C., Ding, J., Pardal, G., Colegrove, P., 2015. Wire+Arc Additive Manufacturing. *Mater. Sci. Technol.* 0, 1–7. doi:10.1179/1743284715Y.0000000073
- Yasa, E., Kempen, K., Kruth, J., 2010. Microstructure and mechanical properties of Maraging Steel 300 after selective laser melting, in: Proceedings of the 21st International Solid Freeform Fabrication Symposium. pp. 383–396

Figure captions

Fig. 1. Experimental setups of the WAAM process. (a) phase 1 (b) phase 2

Fig. 2. Macro-morphology of oxides (phase 1, wire feed speed (WFS) = 1.8m/min, travel speed (TS) = 0.2m/min).

Fig. 3. Micro-morphology of oxides (Acc.V=10kV, Spot=3.0, Det=SE, WD=4.8).

Fig. 4. Cross-sections of oxide layers on each level, thickness of oxide layers and elements distribution (for a-e, Acc.V=10kV, Spot=3.0, Det=SE, WD=4.8). (a) 1 layer (b) 3 layers (c) 5 layers (d) 7 layers (e) 9 layers (f) thickness of oxide layer (g) linear scanning results from Fig. 4d

Fig. 5. Entrapped oxide layer found between layer 6 and layer 7 (a,b) and its SEM elemental mapping results (c,d,e,f). (a) OM (b) SEM (c) O (d) Fe (e) Ti (f) Al

Fig. 6. Footage of deposition process on different surface conditions. (a) on the 2nd layer (on a relatively smoother surface) (b) on the 8th layer (on a relatively rougher surface)

Fig. 7. Wetting and spreading on different surface conditions. (a) on a smooth surface (b) on a rough surface

Fig. 8. Arc voltage variation on different surface conditions.

Fig. 9. Bead shape of WAAM walls in different shielding conditions (phase 2, WFS=8m/min, TS=0.42m/min) (a) torch shielding only (b) torch + tent shielding

Fig. 10. Transverse cross-section of the two walls (from Fig. 9). (a) torch shielding only (b) torch + tent shielding

Fig. 11. Microstructure of the two walls. (a) torch shielding only (b) torch + tent shielding

Fig. 12. Microhardness of the two walls from the bottom to the top.

Fig. 13. Two additional walls built for the tensile test and the tensile specimen drawing. (a) torch shielding only (wall size: 180×68×9.8mm) (b) torch + tent shielding (wall size: 185×68×11.3mm) (c) tensile test specimen drawing

Fig. 14. Tensile test results (error bar: standard deviation).

Fig. 15. Fracture surface of tensile specimens. (a) torch shielding only (b) torch + tent shielding

Table captions

Table 1 Chemical composition of the MARVAL 18S filler wire (wt. %).

Table 2 EDS analysis of marked areas from Fig. 3 (wt. %).

Table 3 Measurements of the two walls.

Table 4 EDS analysis of marked areas from Fig. 15a (wt. %).



DSSPACE

<https://dspace.org/>

Oxide accumulation effects on wire + arc layer-by-layer additive manufacture process

Xu, Xiangfang; Ding, Jialuo; Ganguly, Supriyo; Diao, Chenglei; Williams, Stewart W.

2017-10-20

Elsevier

<http://dx.doi.org/10.1016/j.jmatprotec.2017.10.030>

Xu X, Ding J, Ganguly S, Diao C, Williams S, Oxide accumulation effects on wire + arc layer-by-layer additive manufacture process. Journal of Materials Processing Technology, Volume 252, February 2018, pp. 739-750