

Deformation microstructures and strengthening mechanisms for the wire+arc additively manufactured Al-Mg4.5Mn alloy with inter-layer rolling

Jianglong Gu^{a,*}, Xiaoshu Wang^b, Jing Bai^b, Jialuo Ding^c, Stewart Williams^c,
Yuchun Zhai^b, Kun Liu^d

^aState Key Laboratory of Metastable Materials Science and Technology, Yanshan University,
Qinhuangdao 066004, P.R. China

^bSchool of Materials Science and Engineering, Northeastern University, Shenyang 110819, P.R. China

^cWelding Engineering and Laser Processing Centre, Cranfield University, Bedfordshire MK430AL, UK

^dDepartment of Applied Science, University of Quebec at Chicoutimi, Chicoutimi G7H2W9, Canada

*Corresponding author, Email: gujianglong@ysu.edu.cn, Tel: +86 335 8074780, Fax: +86 335 8514884

Abstract

Applying inter-layer rolling to the wire+arc additively manufacturing (WAAM) process with increasing loads of 15 kN, 30 kN and 45 kN, achieves excellent mechanical properties for 5087 (Al-Mg4.5-Mn) alloys. Compared with the as-deposited alloy, the average micro hardness, yield stress and ultimate tensile strength of 45 kN rolled alloys reached to 107.2 HV, 240 MPa and 344 MPa, which were enhanced by 40%, 69% and 18.2%, respectively. Primary coarse grain structures were found to become greatly refined with an evident rolling texture after deformation. The strengthening mechanisms mainly are deformation strengthening, grain refinement, and solution strengthening. Meanwhile, the elongation of rolled alloys stays over 20%. The plasticity was not obviously diminished compared with the as-deposited alloy. This is two times greater than the commercial wrought Al-Mg alloy with similar composition. The excellent plasticity may be chiefly due to grain refinement, pores closure and reduction, and grain recrystallization during the WAAM re-heating process. The combination process of rolling deformation with WAAM deposition is an effective technique in refining microstructure and improving mechanical properties for AM aluminum alloys.

Key words:

Wire+arc additive manufacturing; Rolling; Aluminum alloy; Microstructure; Mechanical properties

1. Introduction

With recent development in the marine, pressure vessels, military, automotive and aerospace industries [1, 2], non-heat treatable wrought Al-Mg alloys (5xxx) are widely used for large-scale structural components that require low cost, high reliability, good toughness, excellent resistance to corrosion, and relatively high strength-to-weight ratio[3].

Additive manufacturing (AM) of Al-Mg alloy components has attracted more and more attention with advantages in freeform design, materials cost and leading-time saving. There are few research on Al-Mg alloys manufacturing by laser based AM techniques to date. This is because the Mg content reduces the surface tension and increases the melt viscosity, reducing wet ability, thus yielding poor density [5]. Olakanmi et al. [4] reported that an average density of $1.4 \text{ g}\cdot\text{cm}^{-3}$ and a hardness of $85 \text{ kgf}\cdot\text{mm}^{-2}$ for the Al-Mg6 alloy were achieved by selective laser melting (SLM). Beyond this, wire+arc additive manufacturing (WAAM) is another choice for rapid manufacturing of large-scale aluminum alloy parts [6]. Combining of an electric arc as heat source and wire as feedstock, WAAM has been investigated since the 1990s by Cranfield University together with Rolls Royce [7]. Unlimited target parts size, low materials and equipment costs, high deposition and materials utilization rate make WAAM a suitable candidate for replacing the current subtractive manufacturing method from solid billets, especially with regards to low and medium complexity parts [8].

As Colegrove et al. [9] pointed out that one important aspect that is often overlooked for AM parts is that the strength is normally significantly less than that of

commercial wrought alloys. This is the same for WAAM Al-Mg metals, the strength of which is obtained by the insertion of solid-solution element Mg into the Al matrix. However, porosity and large columnar grains are likely to be generated during the arc burning based “micro-casting” of WAAM, causing inferior strength properties of as-deposited alloys. Wang et al. [7] and Ouyang et al. [10] firstly studied the process control and microstructure of variable polarity gas tungsten arc welding (VP-GTAW) based WAAM 5356 (Al-Mg5) alloy, the micro hardness of which is just around 80 HV. Geng et al. [11] found that the average value of ultimate tensile strength (UTS), yield strength (YS) and elongation of the GTAW additively manufactured 5A06 (Al-Mg6.3) alloy are 273 MPa, 124 MPa and 34%, respectively. Apart from WAAM Al-Mg alloys, the maximum UTS for other as-deposited WAAM aluminum alloys, such as 4043 (Al-Si5), 2319 (Al-Cu6.3), are commonly less than 300 MPa [12, 13].

The inferior strength of as-deposited WAAM Al-Mg alloys has impeded their industrial applications. For example, UTS, YS of the conventionally widely-used wrought 5083-H32 (Al-Mg4.5) alloy are 324 MPa and 248 MPa, respectively [14]. This is remarkably 17% and 100% higher than that of the as-deposited WAAM 5A06 aluminum alloy. Therefore, the strength of deposited WAAM aluminum alloys needs to be enhanced to a level similar to wrought alloys for practical industrial applications [15].

Since Al-Mg alloys are non-heat treatable, after-deposition heat treatment strengthening cannot be applied. In the related field of welding, rolling of weld beads is an extremely effective way in improving tensile properties by inducing dislocations [16]

and reducing residual stresses [17]. More recently, two main groups have applied rolling to the WAAM technique with various materials. Colegrove et al. [18] from Cranfield University firstly reported the implementation of inter-layer rolling to the WAAM steel structures deposition in controlling residual stresses and reducing distortions. Rolling was employed between each and every deposited layer at a constant load during the metal deposition process. Martina et al. [20] reported the application of this technique to the GTAW based WAAM Ti-6Al-4V alloy. The UTS and YS of 50 kN and 75 kN rolled deposits are 17% and 20% higher than the unrolled alloy in maximum. Donoghue et al. [21] later identified that the application of a rolling step sequentially to each added layer was surprisingly effective, in resulting in a refined, equiaxed microstructure with a weakened crystallographic texture for the WAAM Ti-6Al-4V alloy. Another research group led by Zhang et al. [19] at Huazhong University from China reported their study on hybrid “in-situ rolling” and deposition. They applied micro rolling directly behind the deposition torch. Xie et al. [22] found that the strength of the WAAM 316L stainless steel could be 33% enhanced with hybrid deposition and rolling.

The authors [15, 23] recently applied the rolling technique to the cold metal transfer (CMT) based WAAM 2219 (Al-Cu6.3) aluminum alloy. They investigated the effect of rolling both on microstructure/mechanical properties improvement and porosity shrinkage. Experimental results show that the micro hardness and strength were gradually enhanced with increasing rolling loads. Moreover, the strength and especially the elongation of rolled materials after heat treatment are much higher than the wrought 2219-T6 alloy. The ability and potential in improving mechanical

properties that can be achieved with rolling suggest that there is likely to be a promising future. More further detailed investigation of benefits of this processing concept on WAAM parts are still being called from the industry. The aim of this work was to study the influence of rolling deformation on microstructures and mechanical properties of the WAAM 5087 (Al-Mg4.5-Mn) alloy. A more profound analysis and description of formation mechanisms are presented.

2. Materials and experiment

Chemical compositions of the commercial ER5087 wire (1.2mm in diameter), the 5083-H32 substrate plate, and the deposited 5087 wall were analyzed by the inductively coupled plasma optical emission spectrometer (ICP-OES) technique with Optima 8300 PerkinElmer, and are listed in Table 1. The dimensions of substrate plates were 300 mm×50 mm×15mm. Before deposition, substrates were washed in the alkaline water and dried in air, which was followed by mechanical cleaning and by degreasing with acetone immediately before usage.

A Fronius CMT Advanced 4000R power source with the pulsed CMT (CMT-P) mode was used for wall building. Compared with the raw wire, composition of the WAAM alloy in Table 1 demonstrates that about 0.32 wt% Mg element was vaporized during arc burning. All the other alloying elements and impurity elements were retained in the deposited alloy. Comparably, there was a 1.2 wt% Mg element lost for the GTAW based WAAM 5A06 aluminum alloy with higher heat input [11]. The small amount of Mg burning loss by CMT suggests that this arc mode is suitable for WAAM deposition.

Table 1 Chemical composition of the ER5087 wire, the 5083 alloy plate, and the

as-deposited (AD) WAAM 5087 wall (The relative error is less than 5% for each tested element.)

Alloys	Chemical compositions (wt%)								
	Si	Fe	Cu	Mn	Mg	Zn	Cr	Ti	Zr
ER5087	0.06	0.16	*	0.74	5.05	*	0.10	0.06	0.10
5083-H32	0.26	0.31	0.06	0.60	4.74	0.08	0.11	0.04	*
AD 5087	0.06	0.16	*	0.73	4.72	*	0.10	0.09	0.13

A rolling rig that was set up by the authors for a previous work [23] was used to move the CMT torch and a flat roller. Multi-layered walls were deposited layer by layer in alternative direction along the centerline of the substrates. Inter-layer rolling was employed between each deposited layer at a constant compressive pressure, which was applied on the top surface of each deposit by a hydraulic cylinder through the roller. With a high pyroconductivity, cooling rate of the aluminum deposit is normally greater than $10^3 \text{ }^\circ\text{C}\cdot\text{s}^{-1}$. Therefore, the practical rolling process is cold working. Parameters for deposition and rolling were all kept constant for all walls as follows: torch/roller travel speed ($0.6 \text{ m}\cdot\text{min}^{-1}$), wire feed speed ($6 \text{ m}\cdot\text{min}^{-1}$), inter-layer cooling time (2 min), shielding gas flow rate ($25 \text{ L}\cdot\text{min}^{-1}$) of pure argon (99.99%), contact tip to work piece distance (15 mm). Besides, heat input during the deposition was maintained around $177.9 \text{ J}\cdot\text{mm}^{-1}$. As-deposited and inter-layer rolled walls with three loads of 15 kN, 30 kN and 45 kN were built. The coordinate system for deposited walls is shown in Fig. 1(a). Wall dimensions and overall deformations – compression rate in the pressing direction – for as-deposited and rolled walls are listed in Table 2.

Both edges (10 mm) of each wall were abandoned. Samples of cross sections parallel to the x-z plane for metallographic observation and hardness tests, were taken

from the center part of each wall. Each specimen was successively ground with 120#, 600#, 1500#, 2500# and 5000# grit SiC papers under flowing water, and polished with 7 μm and 1 μm diamond paste in turn. Colloidal silica suspension (OPS) was applied for final polishing to reach a mirror finish.

Table 2 Dimension and deformation of WAAM 5087 walls

Conditions	Length (mm)	Layers	Total height (mm)	Mean height per layer (mm)	Wall width (mm)	Total deformation (%)
As-deposited	220	30	49.5	1.65	7.5	-
15 kN rolled	220	35	49.6	1.42	8.4	13.9
30 kN rolled	220	45	51	1.13	10.4	31.5
45 kN rolled	220	55	49.5	0.90	13.2	45.5

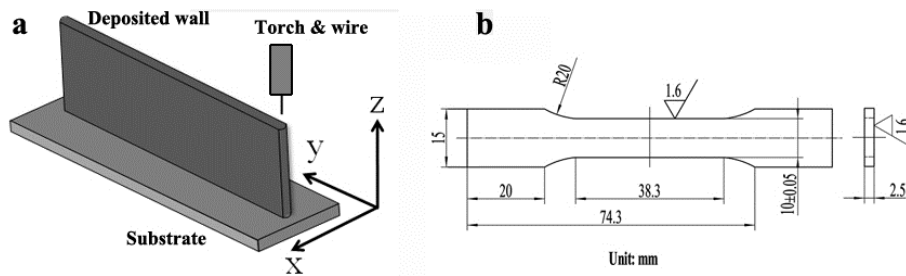


Fig. 1. (a) The coordinate system for the WAAM deposited wall and (b) drawing of the tensile sample

X-ray diffractometer (DX-2500, DANDONG) was used for XRD phase analysis. The scanning angle ranges from 20° to 90° . The microstructures of rolled and unrolled alloys were characterized by using scanning electron microscopy (SEM) (XL30ESEM, PHILIPS). Micro-area composition was measured by energy dispersive spectrometry (EDS). Electron back scattered diffraction (EBSD) maps were performed over an area about $(700-900) \mu\text{m} \times (500-700) \mu\text{m}$ with a step size of $1.5 \mu\text{m}$ by INCA Crystal EBSD system, Oxford Instruments. The Channel 5 software, which is developed by HKL Inc.

(Hobro, Denmark), was used to analyze the data. Specimens for EBSD were polished by direct current with a solution consist of 20 vol% perchloric acid + 80 vol% alcohol at 10 V and 0 °C for 8-15 seconds.

Three tensile test samples parallel to the deposition direction (y axis) were equidistantly sectioned from top to bottom of each wall. Tensile samples as shown in Fig. 1(b) were prepared following the BS EN ISO 6892-1: 2009 standard with a final surface roughness less than 1.6 μm . Tensile tests were processed at room temperature by an electro-mechanical universal testing machine, loading with a constant strain rate of 0.005 min^{-1} . Fracture surfaces morphology of tensile test specimens was observed by SEM. Hardness tests initiated 20 mm below the top surface of each wall. Twenty points with an interval of 0.5 mm were measured along the vertical z direction at the load of 200 g with 15 seconds dwell time by Vickers micro hardness testing machine (Auto-C.A.M.S. of Zwick Roell).

3. Results and discussion

3.1 Mechanical properties

Fig. 2 shows the effect of rolling on micro hardness of WAAM 5087 alloys under increasing loads. The micro hardness values obviously fluctuate ranging along 20 mm on the cross section (x-z plane) of the as-deposited sample. After rolling, micro hardness becomes more and more evenly distributed with increasing loads, indicating gradually uniform mechanical properties after rolling. The mean average micro hardness for the as-deposited, 15 kN rolled, 30 kN rolled and 45 kN rolled samples are 76.6 HV, 87.9 HV, 97.3 HV and 107.2 HV, respectively. Along with increasing rolling loads, average micro

hardness of rolled alloys linearly increased by 14.8%, 27% and 40% compared with the as-deposited alloy. In contrast, the corresponding total deformation rates as listed in Table 2 increased by 13.9%, 31.5% and 45.5%, respectively.

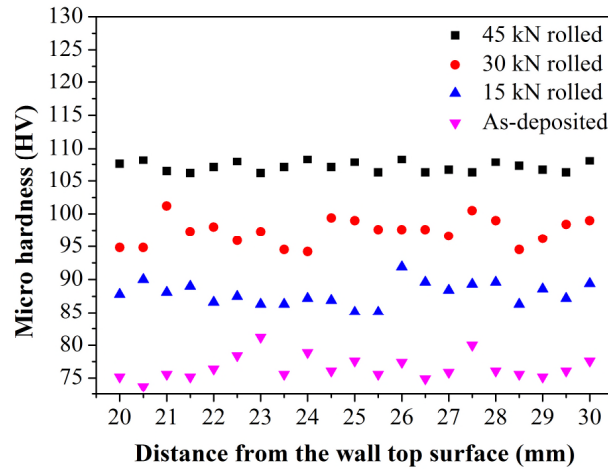


Fig. 2. Micro hardness of as-deposited and rolled WAAM 5087 alloys

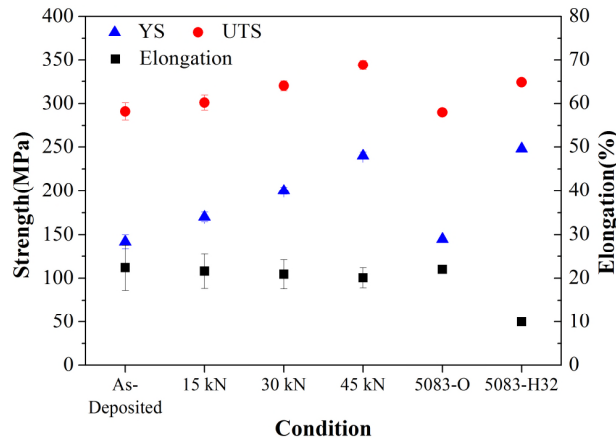


Fig. 3. Tensile properties of as-deposited and rolled WAAM 5087 alloys

Fig. 3 shows tensile properties of YS, UTS and elongation for various conditioned WAAM 5087 alloys and referenced wrought 5083 alloys. The mean YS, UTS and elongation for the as-deposited 5087 alloy are 142 MPa, 291 MPa and 22.4%, which is close to the O-tempered wrought alloy or casting alloys with similar composition. However, these properties are remarkably lower than commonly applied H-tempered wrought alloys. After inter-layer rolling, strength values especially YS significantly

grow with increasing loads and deformation rates. The YS and UTS increase by 19.7% and 3.4%, 40.8% and 10%, 69% and 18.2%, respectively for 15 kN, 30 kN and 45 kN rolled WAAM alloys. The allowable strength of this WAAM alloy was remarkably improved by rolling. Furthermore, along with increasing rolling loads, the gradually shorter error bars imply that rolling improves homogeneity of WAAM alloys mechanical properties.

Another noteworthy characteristic is the nearly undiminished plasticity of rolled alloys. The elongation of the as-deposited and rolled alloys are 22.4%, 21.6%, 20.9% and 20.1%, respectively. In addition, elongation value of the 45 kN rolled WAAM 5087 alloy is two times greater than the commercial 5083-H32 alloy. As Wang et al. [24] reported, the elongation of the rolled wrought 5052 (Al-Mg2.5) alloy under 46% reduction is just about 5%. This indicates that there may be wider application areas of this investigated alloy. Reasons for strengthening and plasticity maintaining ability of rolled WAAM Al-Mg alloys are analyzed in Section 3.3.

Apart from the notably excellent plasticity, YS and UTS of the 45 kN rolled alloy reach to 240 MPa and 344 MPa, respectively. These values are close to or higher than standard commercial Al-Mg alloys with similar compositions, implying that the investigated alloy could meet the industrial requirements for practical implementation.

3.2 Fracture analysis

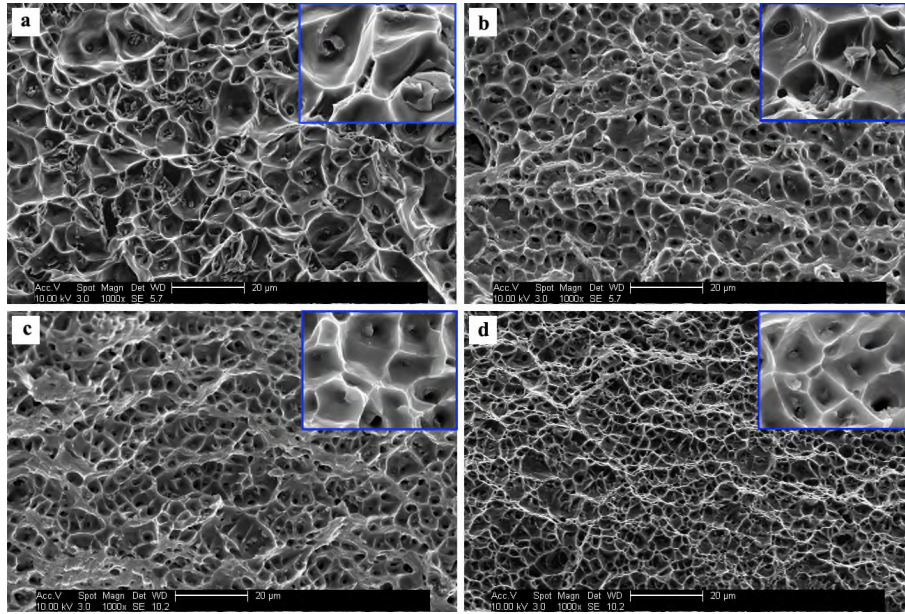


Fig. 4. SEM fractograph for tensile test specimens of WAAM 5087 alloys (a)
as-deposited; (b-d) 15 kN, 30 kN, 45 kN inter-layer rolled

Typical SEM images of fractured surfaces of different conditioned samples after tensile tests show various dimples sizes in Fig. 4. Coarse primary dimples shown in Fig. 4(a) for the as-deposited alloy indicate ductile mode fracture leading to high tensile elongation. This is because those characteristics of ductile fracture are supposed to be tearing of metal accompanied by considerable plastic deformation. It is evident in Figs. 4(b-d) that dimples for the rolled samples become finer and more uniform along with increasing rolling loads. Formation of finer and denser dimples, which commonly imply higher strength for ductile metals, may be attributed to nucleation of a large number of micro voids due to severe strain induced during deformation. Moreover, fractured surface of the as-deposited alloy in Fig. 4(a) shows dimples embedded with coarser and cracked second phase particles, which may diminish the tensile strength. This is because for ductile materials, a crack is formed by coalescence of micro voids that may form as a result of cracking of second phase particles [25]. Actually, a fragmentation of Fe and

Mn bearing particles was observed for the rolled samples by SEM. EDS analysis of those phase particles and effects of rolling on them are observed and discussed in Section 3.3.

3.3 Microstructure and strengthening mechanisms

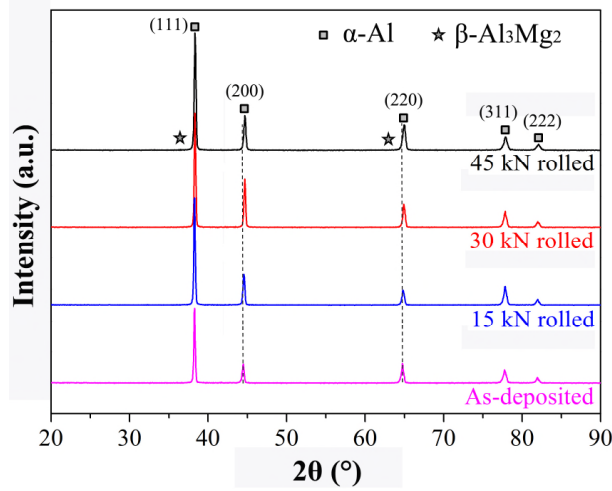


Fig. 5. X-ray diffraction spectra for as-deposited and rolled WAAM 5087 alloys

As shown in Fig. 5. XRD results of WAAM 5087 alloys, new diffraction peaks are not detected after rolling with increasing loads, indicating that phase transformations were not occurred during the hybrid process. Obvious differences among the four states lie in the α -Al phase. There is a fairly strong peak of the main (111) crystallographic orientation, the peak intensity of which increases obviously along with increasing rolling loads. Small variations of α -Al phase intensity indicate the percentage of this particular orientation vary from one with the others. This may be caused by the lattice plane rotation under deformation. Furthermore, diffraction spectra for (200) and (220) orientations of α -Al are shifted right with increasing loads. According to the Bragg equation, larger diffraction angle was caused by smaller lattice parameters. Those compressed plane orientations maybe the main deformation planes under rolling.

SEM images for cross sections of WAAM 5087 alloys are shown in Fig. 6. Two main features can be drawn from the randomly distributed pores and second phase particles within the alloys. One is that the number and size of black round-shaped pores are gradually reduced under increasing rolling loads, reasons of which were explained in a previous study by the authors [23]. The closure and reduction of pores after rolling may be one possible reason for remarkable strength enhancement without plasticity decrease. This is because as Kobayashi et al. [26] pointed out, porosity is harmful to the strength of aluminum alloys. Porous regions probably yield first due to the reduced tensile load bearing capacity, causing concentration of strains near the voids thus premature fracture [27].

Another distinct feature shown in Fig. 6 is the finer and more evenly distribution of white second phases with increasing rolling loads. As shown in Fig. 6(d) with a higher magnification, these white particles were fractured into smaller pieces after rolling. EDS analysis in Fig. 7 reveals that homogeneously distributed coarse particles contain Al, Mn and Fe elements. According to ref. [28], those light coarse particles dominated in grains, were identified as $\text{Al}_6(\text{Mn,Fe})$. Other particles of Al-Mg, Al-Ti and Mg-Si were also found, although they were not detected with the XRD analysis due to their low contents. Spectrum A on the rod-like phase and spectrum D on the particle-like phase contain Ti and Si elements, respectively. Those particles may be the trace Al_3Ti or Mg_2Si doped $\text{Al}_6(\text{Mn, Fe})$ phases. Further investigation will be undertaken to investigate the influence of those particles on mechanical properties of this alloy.

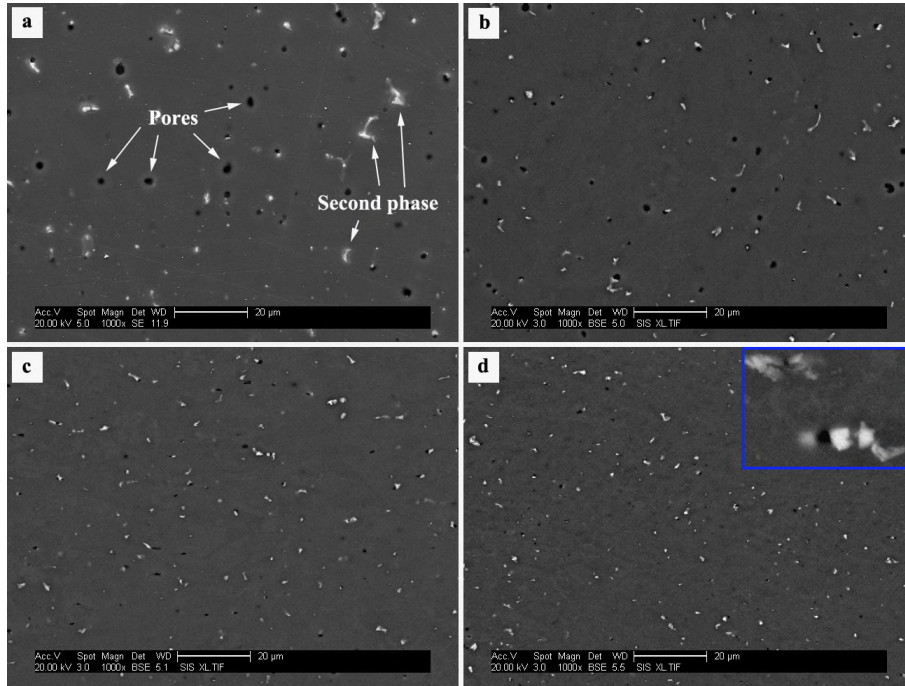


Fig. 6. SEM images for WAAM 5087 alloys in states of (a) as-deposited; (b-d) 15 kN, 30 kN, 45 kN inter-layer rolled

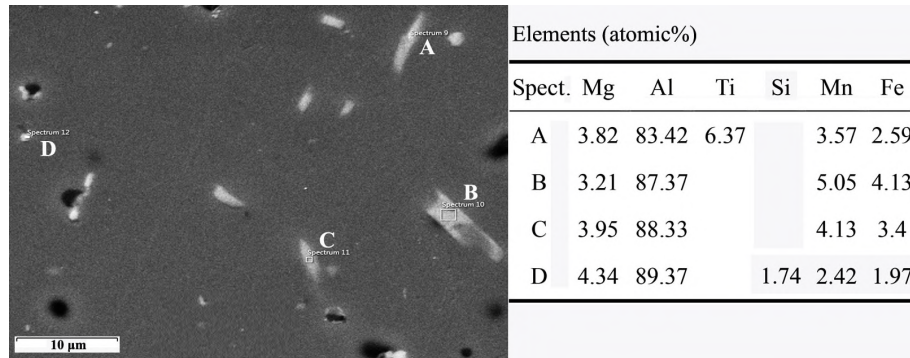


Fig. 7. EDS spectra of particles in the as-deposited WAAM 5087 alloy

After cold working, accumulated strains generated by plastic deformation result in high-density dislocations piling up and substantial tangling, exerting significant obstacle effect on dislocation movements and prompting resistance to deformations. Therefore, mechanical properties of WAAM alloys would be improved by the deformation strengthening mechanism after rolling, accompanying with distinct microstructures. The grain orientation, texture, grain sizes, and boundary misorientation angle distribution

information were analyzed with EBSD and the CHANNEL 5 software. Coloring inverse pole figures (IPF) from cross sections (x-z plane) of as-deposited and rolled samples are depicted in Fig. 8. Microstructure of the as-deposited metal consists of primary equiaxed grains without texture. Although grain morphology of the 15 kN rolled sample was not greatly changed, low misorientation shows up in the grain interior regions. In contrast, greatly refined equiaxed grain structures with a very weak texture were observed when the load of 30 kN was applied. Grains were further refined after rolling at 45 kN with higher deformation rate.

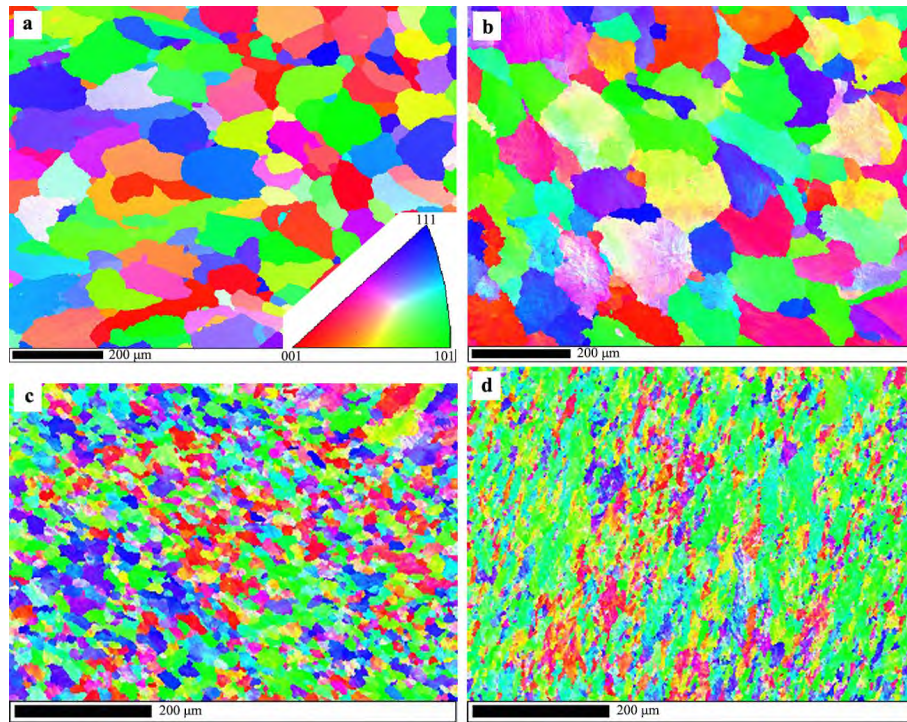


Fig. 8. IPF coloring orientation maps for WAAM 5087 alloys (a) as-deposited and coloring legend; (b-d) 15kN, 30kN, 45kN inter-layer rolled

Pole figures of various conditioned α -phase aluminum texture are shown in Fig. 9. There is a random texture in the as deposited and 15 kN rolled walls with a maximum intensity of less than approximately 3.5 times above the random background, though a

weak cube texture $\{001\}\langle 100\rangle$ is observed in the 100 pole figure of Fig. 9(a). Much stronger texture was found in the 30 kN and 45 kN rolled samples. As can be seen from Fig. 9(c) and (d) both the 30 kN and 45 kN rolled walls' 111 pole figures, rolling texture components including brass $\{110\}\langle 1\bar{1}2\rangle$, S $\{123\}\langle 634\rangle$ become evident. With greater rolling force, the maximum intensity increasing from 5.01 times random for the 30 kN load to 5.68 for the 45 kN load.

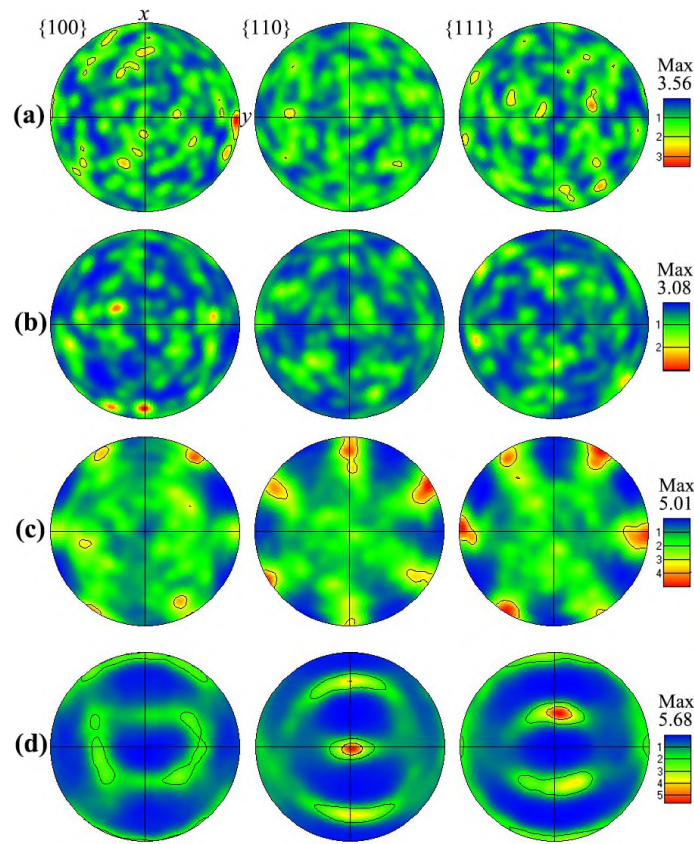


Fig. 9. Pole figures for WAAM 5087 alloys (a) as-deposited and coloring legend;
(b-d) 15kN, 30kN, 45kN inter-layer rolled

Compared with ideal texture, the texture components with a deviation $<15^\circ$ for WAAM 5087 walls under different rolling loads were calculated by the CHANNEL 5 software and listed in Table 3. The dominating texture divides into two types, one which is recrystallization texture of cube $\{001\}\langle 100\rangle$ and goss $\{110\}\langle 001\rangle$, one which is

rolling texture of brass $\{110\}\langle 1\bar{1}2\rangle$, S $\{123\}\langle 63\bar{4}\rangle$ and copper $\{112\}\langle 11\bar{1}\rangle$. Along with the increasing force, the total amount of rolling texture components increased gradually from 20.95% to 60.33%, indicating that the deformation texture is in major after 45 kN rolling. The brass and S components each evidently increased from around 6% to over 20% under 45 kN rolling, while the change is not obvious for the copper component. The quantity of goss and cube components irregularly varied from as deposition to 45 kN rolling. This may be caused by the complex cycling heat input-cold work process.

Table 3 Texture components (%) for WAAM 5087 walls under different loads

Conditions	Brass	S	Copper	Goss	Cube	Random	Total rolling texture
As-deposited	8.08	5.92	8.58	7.38	3.65	66.39	22.58
15 kN rolled	6.09	6.42	8.44	9.12	4.19	65.74	20.95
30 kN rolled	11.19	10.4	13.7	6.4	4.17	54.14	35.29
45 kN rolled	25.51	21.7	13.12	14.63	1.84	23.2	60.33

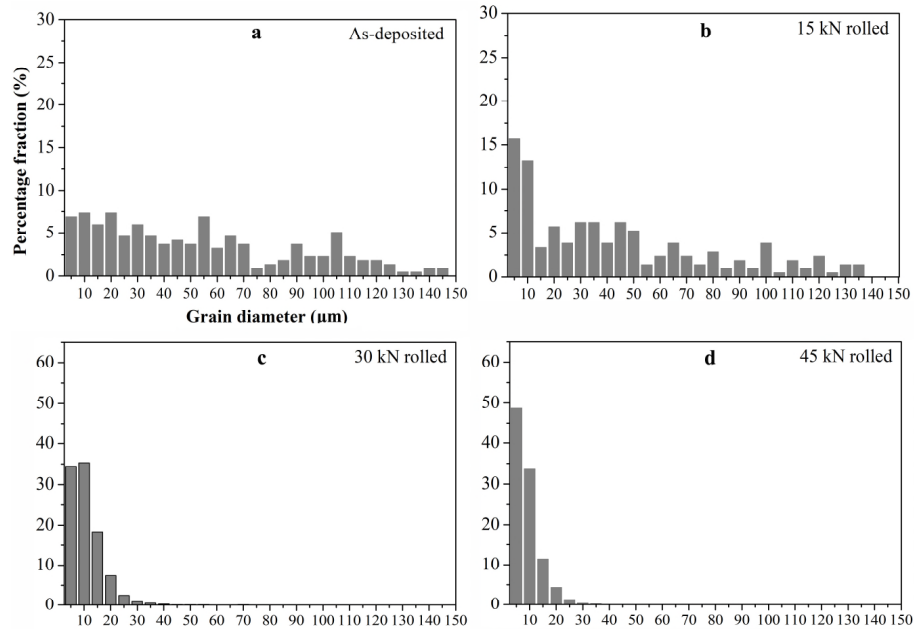


Fig. 10. Grain diameters distribution of WAAM 5087 alloys (a) as-deposited; (b-d) 15 kN, 30 kN, 45 kN inter-layer rolled

It is evident that grains were greatly refined after inter-layer rolling, while they were not greatly elongated along the rolling or the metal deformation direction. This may be caused by combined action of cold working and continuous subsequent heat input. Grains recrystallization and refinement may thus occur due to the re-heating of rolled structures due to subsequent deposition.

Grain size is one of the most important micro structural factors in controlling mechanical properties for aluminum alloys. If grain or sub-grain sizes are to be determined from an EBSD map then consideration must be given to the pixel step size in relation to the grain size. According to Humphreys criterion [29], 5 pixels (1 in 4) per grain are required to obtain an accuracy of 10%. This means there should be at least 3 pixels in a row within a grain or subgrain region. Therefore, grains larger than 4.5 μm were counted for this research due to the limitation of EBSD scanning resolution. Histograms in Fig. 10 show the grain sizes distribution by sorting in diameters with an interval of 5 μm . As can be seen, Grain sizes become gradually smaller with increased rolling loads, indicating a grain refinement strengthening effect. The average grain sizes were found to be reduced from 53.9 μm to 45.7 μm by applying a 15 kN load, and were further reduced to 11.0 μm and 9.1 μm under loads of 30 kN and 45 kN, respectively.

In this research, grains were defined as fine grains (FG), small grains (SG), and coarse grains (CG) according to three diameter scales of $<10\text{ }\mu\text{m}$, $10\text{ }\mu\text{m}$ - $30\text{ }\mu\text{m}$, and $>30\text{ }\mu\text{m}$, respectively. As shown in Fig. 10, the proportion of FG reaches to 28.9% after 15 kN rolling from 14.2% without rolling. There is an obvious decrease of CG from 61.9% to 52.1% after 15 kN rolling. Along with increasing rolling loads, FG scales

under 30 kN and 45 kN continuously increase to 69.7% and 82.2%, respectively. SG grains take up 23.9%, 19.0%, 29.1% and 17.4% respectively for the as deposited and the increasing rolled alloys. Most notably, the percentage of CG under 30 kN is 1.2%, which is greatly reduced compared with the alloy under 15 kN rolling. Correspondingly, there are only 0.4% CG grains in the 45 kN rolled alloy. The total proportion values of FG and SG grains for the as-deposited, 15kN, 30 kN and 45 kN rolled alloys are 38.1%, 47.9%, 98.8%, 99.6%, respectively. It can be concluded from the above analysis that the hybrid process of cold working and WAAM, especially under loads higher than 30 kN, is benefit in grain refinement for the Al-Mg alloy. This process can significantly decrease the CG proportion and increase the FG proportion, which could effectively improve the strength properties while maintaining excellent plasticity properties for this Al-Mg alloy.

This indicates that grain refinement may be the main strengthening mechanism for the WAAM 5087 aluminum alloy, because it is well known that mechanical properties of a metal are strongly influenced by the grain and sub-grain sizes. The Hall-Petch equation depicts an increase in yield stress (σ_s) with a decrease in grain size (d) [30, 31]

$$\sigma_s = \sigma_0 + kd^{1/2} \quad (1)$$

Where σ_0 and k are constants determined experimentally. The relations between grain sizes $d^{1/2}$ and yield stresses of rolled and unrolled alloys are plotted in Fig. 11, indicating that the strength growth fits well with the decreasing grain sizes. The corresponding Petch parameters are $\sigma_0=102.3\text{MPa}$ and $k=384\text{MPa}\cdot\mu\text{m}^{-1/2}$ with a slope error of 8%.

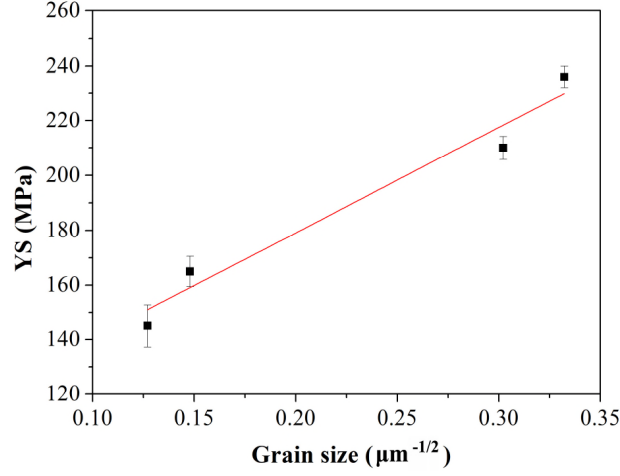


Fig. 11. Relationship between the yield stress and the grain size $d^{-1/2}$

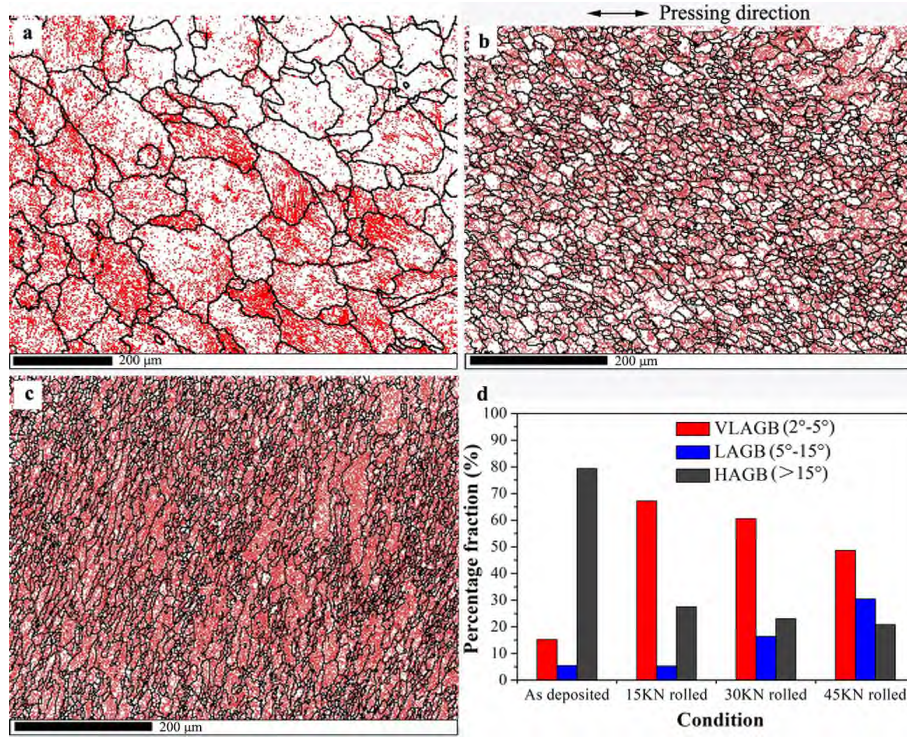


Fig. 12. Typical OIM micrographs of WAAM 5087 alloys (a-c) 15 kN, 30 kN, 45 kN rolled correspondingly. Red lines correspond to boundaries with low misorientation $\theta < 15^\circ$, black lines $\theta > 15^\circ$. (d) misorientation angle distribution of as-deposited and rolled WAAM 5087 alloys

When a load of 45 kN was applied, around 50% grains were compressed to around 5 μm . This is because a great amount of finer sub-grains are formed after rolling by

splitting primary grains of the deposited alloy, inducing low misorientation angle grains. Low misorientation is an indication of sub-grain boundaries, which is formed due to cell walls generated by dislocation climbing, sliding and tangling [15]. Fig. 12(d) shows the grain boundary misorientation of WAAM 5087 alloys. Grain boundaries with misorientations larger than 15° were defined as high angle grain boundaries (HAGB). Those with lower misorientations between 5° - 15° were defined as low angle grain boundaries (LAGB), and those with misorientations between 2° - 5° as very low angle grain boundaries (VLGB).

According to Fig. 12(d), fraction of HAGB accounts for around 80% of all the boundaries, indicating a highly crystallized state for the as-deposited alloy. When the WAAM deposited metal was cold rolled after each layer, the fraction of low angle grain boundaries obviously increased. This indicates that along with the increased deformation strains, a large amount of sub-grains were formed, demonstrating severe grain splitting. This high density of sub-grains integrated with large scale of FG caused by grain fragmentation, was developed into misoriented regions in original grain interiors. After 15 kN rolling, the percentage of VLGB increased to 67.2% from 15.2% as-deposited, indicating a positive recovery for the rolled metal due to the subsequent reheating processes. Along with increasing rolling loads, the proportion of VLGB decreased to 60.5% and 48.7% under 30 kN and 45 kN, respectively. While the LAGB stays unchanged for the 15 kN rolled alloy. The scales of LAGB under 30 kN and 45 kN rised to 16.4% and 30.4% respectively. This may be caused by the temperature gradient within the deformed metal when heated up by subsequent

depositions. With higher rolling loads, the layer height gets smaller, indicating each deformed layer becomes closer to the heat source, i.e. the arc. Recrystallization would be more easily occurred on those sub-grains with higher temperature.

Typical orientation imaging microscopy (OIM) graphs of the rolled microstructures are shown in Fig. 12(a-c). Grain boundaries were colored to highlight the size distribution of the refined grains. Orientation differences between neighboring grains, $\theta < 15^\circ$ or $\theta > 15^\circ$, are separately marked by red lines and black lines. New fine grains are evolved with further deformation under higher loads, while coarse grains with high angles still exist. As shown in Fig. 12(a) for the stage of 15 kN rolling, large primary grains was not obviously reduced, and low grain boundaries are inhomogeneously developed accompanied with embryos of dislocation substructures. In other words, this is an incubation stage for formation of FG grains. Moderate stains appear under 30 kN rolling. Extension of large grains could be observed, accompanying with great deformation inside of grains.

With further rolling deformation under 45 kN, grains with high misorientations were elongated in the freeform deformation (x) direction, which is vertical to the pressing direction. Weak fibrous microstructure consisting of small deformation bands were thus found aligned to the pressing direction. Although the percentage of HAGB was reduced, the practical amount of grains in this type was increased. The recrystallized grains may be generated due to the combining effect of cold working and subsequent thermal action. This is because dynamic recovery can assist the transformation of strain-induced non-equilibrium boundaries to equilibrium ones,

forming new grains during strain deformation with thermal functions [32]. In addition, the new and high density of low misorientation can act as nuclei after re-heating. New fine grains with low angle dislocation sub-grain boundaries are homogeneously developed in the deformed HAGB grain interiors.

In summary, the strain-induced grains evolvement after rolling for WAAM 5087 alloys suggests that new grains, both large and small ones, may be mainly controlled by grains deformation, grain fragmentation, and recrystallization. Therefore, alloys were enhanced due to significant grain refinement strengthening.

There are some other presumed strengthening mechanisms for the deformed Al-Mg alloy due to alloying elements addition. Wei et al. [33] pointed out that the mechanical properties of Al-Mg alloys could be improved by solution strengthening. The pressing load of rolling increased the solubility of Mg atoms in the Al matrix. The solid-soluted Mg atom distorted the lattice of matrix alloy. Consequently, the resistance of dislocation movement was increased and led to solution strengthening. Moreover, as Radović et al. [34] reported that the addition alloying elements would increase both size and volume fraction of the dispersoids of transition metal particles, such as Al_6Mn , Al_3Ti . Such particles could effectively pin the grain boundaries, playing an important role for retardation of dislocation rearrangements [35]. For the hybrid manufactured 5087 aluminum alloy, the fractured Mn, Ti etc. bearing phase particles could improve stability of strain induced new grain structures, in leading to suppression of grain growth of recrystallized grains during deformation for this Al-Mg-Mn alloy.

The plasticity of WAAM 5087 alloys was not obviously diminished compared with

the as-deposited alloy by the inter-layer rolling process. The first reason for this is because of the grain refinement effect, which and sub-grain formation can contribute to increase in strength without significant loss in ductility. Secondly, the loss in plasticity due to high strain deformation has been compensated by occurrence of recrystallization during the WAAM re-heating process. In addition, the low intensity of coarse second phase particles as shown in Fig. 6 exerts low impact on plasticity properties. Especially under higher rolling loads, those particles were fractured and refined, becoming more evenly distributed. The strength and plasticity of Al matrix would be both improved rather than closed to a brittle fracture. Consequently, the combination of rolling deformation with WAAM deposition process could be a potential technique in refining microstructure and improving mechanical properties for aluminum alloys.

4. Conclusions

Key findings that can be drawn from the study on effects of rolling on microstructures and mechanical properties of WAAM 5087 alloys are as following:

- (1) When each deposited layer was sequentially rolled during the WAAM process, the average YS, UTS and micro hardness grew linearly along with increasing loads of 15 kN, 30 kN and 45 kN. Meanwhile, the elongation of rolled alloys stays nearly the same as the as-deposited alloy. All rolled alloys were fractured in the ductile mode with a distinct feature of fine dimples.
- (2) Primary coarse grain structures were found to become greatly refined with an evident rolling texture after deformation. Under 45 kN load, fine grains less than 10 μm account for 82.2% of all the grains. Large amount of low angle

grains were found generated in the grain interior regions.

- (3) Strengthening mechanisms for rolled alloys, especially under 45 kN load, were mainly led by deformation induced high density dislocations and sub-structures, and by grain refinement. A Hall-Petch model was established.
- (4) Grain refinement together with pores closure and reduction are the main reasons for the excellent plasticity of rolled WAAM alloys. Beyond these, the loss in plasticity due to high strain deformation has been compensated by occurrence of recrystallization during the WAAM re-heating process. In addition, the low intensity of coarse second phase particles exerts a low impact on mechanical properties.

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Gu, Jianglong; Wang, Xiaoshu; Bai, Jing; Ding, Jialuo; Williams, Stewart W. ...

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