The Effect of In Process Heat Treatment on the Mechanical Properties of Cold Spray Coatings

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Abstract

Cold spray (CS) is an additive manufacturing process which has recently gained interest within research and select high-end production systems; however, one of the primary factors limiting the deployment of this process is the relatively poor mechanical properties of the coatings in the as-sprayed state as compared with bulk counterparts. Therefore, this paper details the mechanical performance of Al 6061 CS coatings heat treated using a novel focused IR device. The heat treatment of the coatings was performed in-process with the aim of improving the ductility and strength of the CSed coatings processed using nitrogen carrier gas. The mechanical properties of the heat treated samples were compared to a traditional annealing process and an as-sprayed sample by tensile testing dog bone samples. It was found that the rapid IR heat treatment process delivered advantages with regard to mechanical properties when compared to the untreated state showing an increase in the UTS of the coatings by 52% and an increase in the elongation at failure by 43%. This work demonstrates that rapid heat treatment can be carried out on a CS coating, allowing targeted improvement of mechanical properties over load-bearing components in a more time-effective way than traditional heat treatment techniques allow.

Introduction

CS is an additive manufacturing technique where spherical metallic powders with diameters in the range of 10-50 µm are accelerated to speeds of 300-1200 m/s using a De Laval nozzle before impacting upon a substrate [1,2]. A schematic of the CS process is illustrated in Fig. 1 showing the De Laval nozzle, power injection and coating formation upon a substrate. The kinetic energy of the particle upon impact causes substantial plastic deformation of both the particle and substrate allowing substrate-coating and coating-coating bonding to occur through mechanical interlocking and adiabatic shear instability [2,3]. CS allows coatings to be manufactured which exhibit little oxide inclusion, low porosity and are free from melting and hence significant adverse changes in microstructure. These characteristics allow the manufacture of coatings with microstructural properties which are closer to those of the feedstock powder as compared to alternative thermal spray processes [1]. CS has been successfully applied to the deposition of a wide variety of materials and substrates for different applications including aluminium and its alloys [4–11].

Currently, there are a number of limitations associated with CS, particularly when employed within load bearing applications. Firstly, CS samples are typically significantly more brittle when compared to bulk counterparts due to the strain hardening which occurs during deposition. This greatly affects the fatigue strength of the coatings and their capability to be employed in mechanically loaded applications. Additionally, CS relies on the deformation of the impacting particles and substrate in order to form mechanical and metallurgical bonds; however, materials which deliver a high strength-to-weight ratio are less likely to deform during the deposition process and therefore typically result in lower interparticle bond strength. Therefore, heat treatment processes are often employed to negate defects, improve interparticle bond strength and overall mechanical properties of CSed aluminium coatings [7,12] and other materials [13,14]

However, a full anneal or stress relieving process requires significant processing time due to the large bulk which must be heated, held at a specified temperature (or

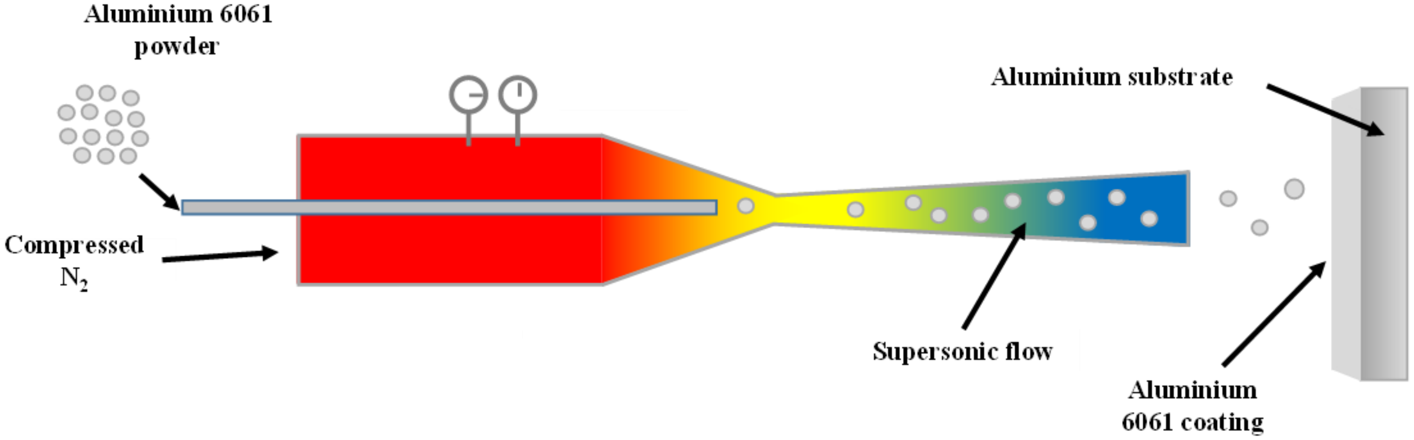


Figure 1: Cold spray schematic

temperatures), and subsequently cooled in order for a single heat treatment cycle. In addition, heat treatment of a whole component is in some cases not possible, for example, when heat treating a component repaired by cold spray with dissimilar materials. Recently, studies have suggested laser annealing as a localised heat treatment process which can potentially improve the mechanical properties of coatings [15,16] and remove work hardening from an Al alloy [17] which is known to increase the ductility of the sprayed material. Laser annealing, and a similar technique using focused IR radiation, were investigated with CS coatings and were shown to decrease the hardness of the materials [18,19]. These rapid heat treatment techniques quickly increase the coating to high temperatures but below that of the melting temperature for a short period of the order of minutes. This method has the additional benefit that the heat treatment can be targeted over the load bearing area of the component and, therefore, not all of the component requires treatment. Hence, this technique is particularly suitable for the manufacture of large components or the repair of damaged components using the CS method. Therefore, this study will expand upon the previous work of the authors to determine the effect of in-process rapid heat treatment on the mechanical properties of hardness, strength and ductility of coatings manufactured using CS.

Experimental setup

Coating fabrication procedure

The coatings were manufactured using the in-house CS system located at Trinity College Dublin. The CS system consists of a de Laval nozzle, gas heater, CNC system, powder feeder, computer monitoring and control system, and a gas supply. Nitrogen was used as the process gas for all the experiments undertaken in this research at a single gas heating temperature of 700ºC. Refer to Table 1 for all of the cold spray processing parameter used in this research. The system was allowed to stabilise for 1 minute ensuring the gas heater reached equilibrium before spraying commenced. The gas and entrained powder enter the converging-diverging de Laval nozzle which accelerates the powders to supersonic speeds. High purity aluminium 6061 (A6061-40A3 Toyal, Japan) was used which is specified by the manufacturer to have a d_{10}, d_{50} and d_{90} of 33.2, 48.4 and 74.6 μm respectively. A nozzle cooling jacket, similar to the device used by Wang et al. [20], was employed to prevent nozzle clogging and allow the spraying of Aluminium at high temperatures. The coatings were sprayed onto a 2 mm thick aluminium 5 series alloy substrate after cleaning of the substrate surface with isopropyl alcohol. All samples were sprayed at a traverse speed of 200 mm/s and a standoff distance of 40 mm. A bidirectional spray strategy was employed with a hatch distance of 5 mm. The second layer was offset by 2.5 mm such that each layer was sprayed in between the two hatches of the previous. The final thickness of each coating was between 6 and 7 mm for a total spray time of approximately 7 minutes.

Heat treatment procedure

A schematic of the manufacturing and heat treatment processes are shown in Fig. 2. A rapid heat treatment was carried out using a focused infrared heater similar to the unit described in the author's previous work [19]. This device uses an optimised reflector shape to concentrates broadband IR radiation emanating from a quartz tungsten halogen (QTH) lamp which can then be used to heat a surface in a more cost-effective manner than a medium to high powered laser. A sectioned view of the reflector is shown in Fig. 2b. The power level of the unit was set to 130 W emanating from a 10 mm diameter exit located 1-3 mm above the surface being treated. The heat treatment was carried out in process at two speeds 10 and 40 mm/s after every layer deposited by the CS following the same path as the CS nozzle. The total time for the heat treatment process was 35 and 140 minutes for the 40 mm/s and 10 mm/s samples, respectively. The CS powder flow was paused during the heat treatment and reinitialised after the heat treatment on that layer concluded. The rapidly heat treated samples were compared to a traditional full annealing heat treatment (3 hours at 410ºC, cooled at 40 ºC/ per hour to 260ºC followed by air-cooling [21]) with a ramp rate of 10ºC/min. A shorter stress relieving heat treatment process (1 hour at 350ºC) was also tested as an intermediate heat treatment which may be more comparable to the rapid heat treatment process. The samples were allowed to air cool after heat treatment within the furnace. The heat treatment process was carried out before the manufacture of dog bone shapes.

Characterisation of coating microstructure

To carry out analysis of the coating cross sections, the samples were polished using standard metallographic procedures with the final polishing applied using 0.06 μm colloidal silica solution. The microscope images were obtained at a magnification of 200x and 500x. Etching was carried out by exposing polished surfaces to Keller's reagent for approximately 40 seconds allowing interparticle and grain boundaries to be inspected and observe microstructural changes due to heat treatments. The micro-hardness for each mounted coating was determined using a Vickers hardness machine (MVK-H1, Mitutoyo, Japan) using an average of at least 5 measurements with a 25 g load and 10 second dwell time.

Mechanical testing procedure

The CS samples were removed from the substrate and cut into 5 dog bone samples using a wire EDM (Excetek V400G,

Table 1: cold spray processing parameters

Process gas	Inlet pressure	Gas temperature	Traverse speed	Standoff distance	Powder feed rate
Nitrogen	3 MPa	700ºC	200 mm/s	40 mm	60 g/min

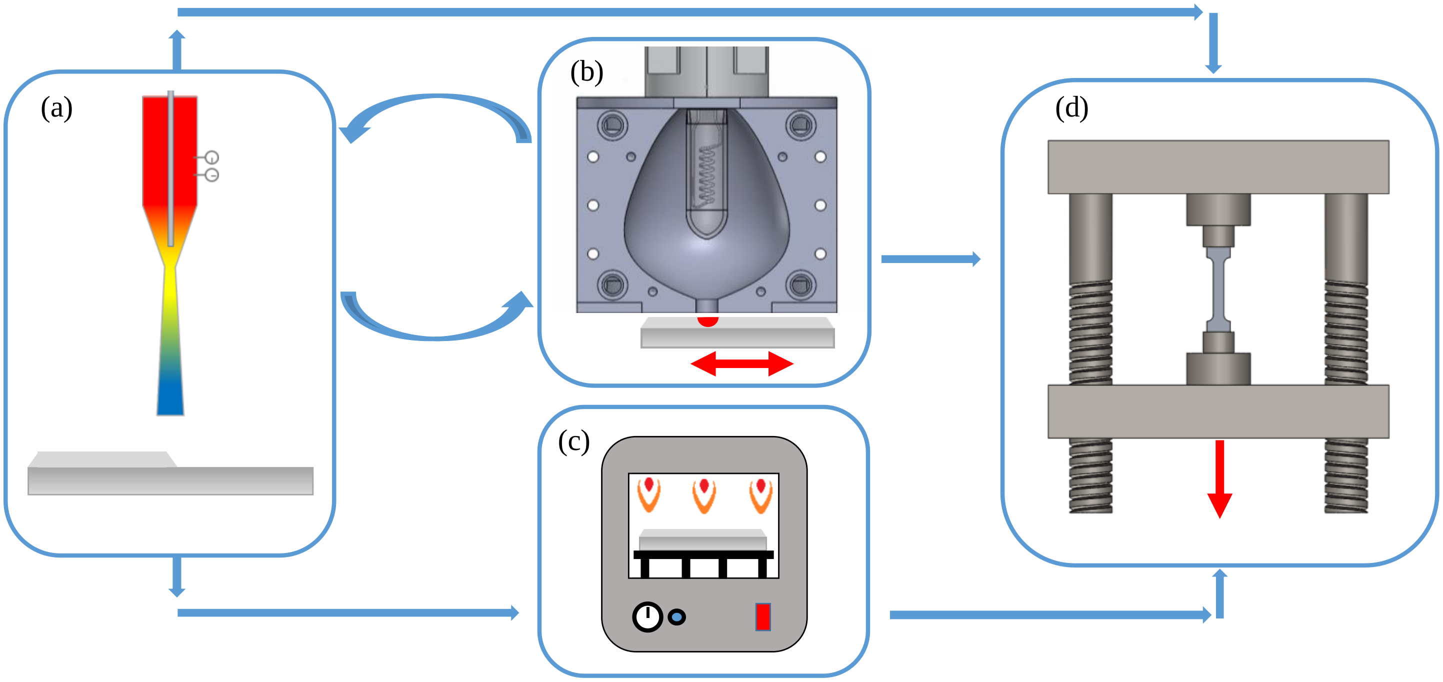


Figure 2: Manufacturing process schematic with: (a) the cold spray coating manufacture, (b) rapid IR heat treatment, (c) traditional heat treatment, (d) tensile testing of dog bone samples

Taiwan). The tensile test samples were cut to a gauge length of 25 mm, a width of 5 mm, a thickness of 4 mm and an overall length of 54 mm. The ultimate tensile stress (UTS) and elongation at failure (EL) of the deposits were determined based on the average value of at least three of tensile specimens. A number of the weaker dog bone samples failed during tensile test setup and were therefore omitted from the provided data. Tensile testing was carried out using a universal tensile strength system (Instron 3366, UK) at a displacement rate of 1 mm/min.

Results and discussion

The effect of the heat treatment on the micro-hardness of the samples is presented in Fig. 3 and shows a trend of reduction in hardness with an increase in heat treatment time. The as-sprayed sample is the second hardest of all the specimens tested only exceeded by the 40 mm/s rapid heat treatment test. However, this is within the uncertainty of the hardness measurement, and it would be expected that the as-sprayed sample would be the hardest if the measurement uncertainty was reduced. The as-sprayed sample with an average Hv of 60.1 is significantly harder than the bulk of Al 6061 T0 which was measured to be 34 Hv. This is due to the work hardening of the sample which occurs during deposition and the uncontrolled cooling during powder formation. The fully annealed sample delivered the lowest micro-hardness of all the specimens tested, as expected, with a 49% reduction when compared to the as-sprayed sample. The stress relieved sample, which has a much shorter processing time than a full anneal, also shows a significant reduction in hardness by 44% when compared to the as sprayed sample. The rapid heat treatment process at 10 mm/s delivered a 14% reduction in hardness. The 40 mm/s test did not have a measurable effect on the hardness of the sample based on the tests performed.

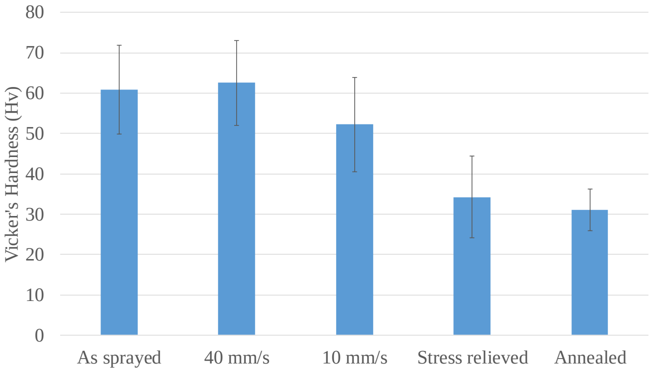


Figure 3: The effect of various heat treatments on the hardness of the CSed Al 6061 samples.

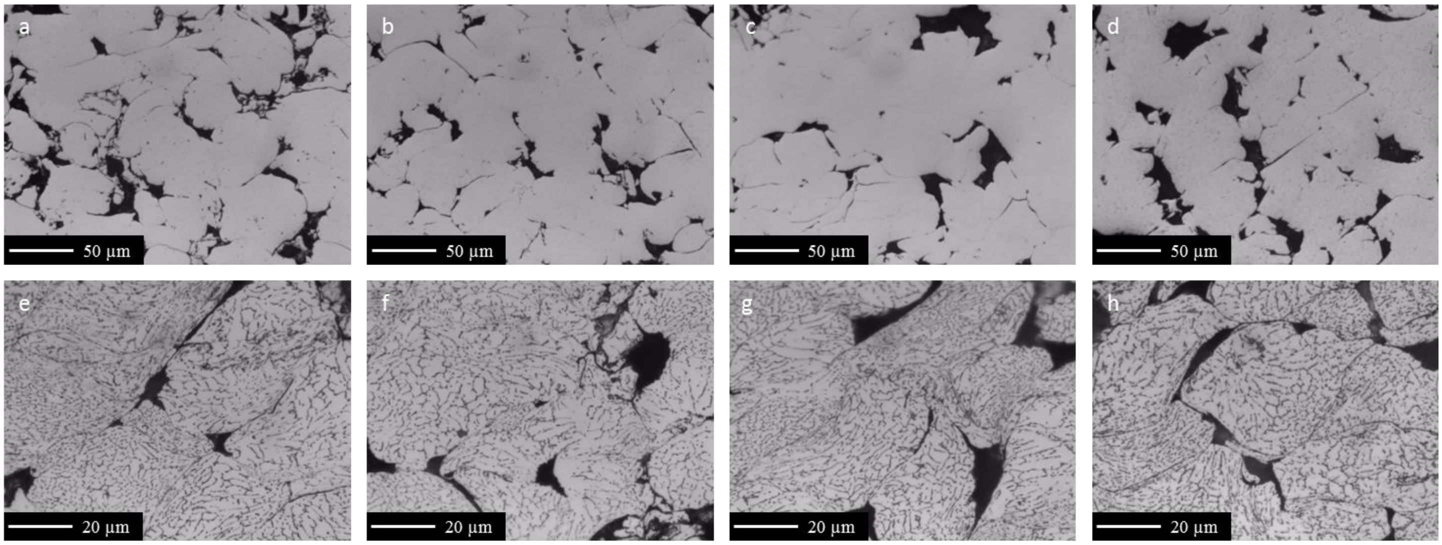


Figure 4: Cross sections of the microstructure of the plain and etched of the as sprayed (a,e), 40 mm/s rapid heat treatment (b,f), 10 mm/s rapid heat treatment (c,g), and the fully annealed sample (d,h)

Statistically, it is not conclusive with this sample if the rapid heat treatment had a significant effect on the hardness of the coatings, although, on average, the data suggests that the 10 mm/s test likely reduced the coating hardness. Further testing is required for conclusive evidence. The 40 mm/s traverse speed may be too high to cause a measurable effect using this technique, and therefore a higher power or slower speeds must be used in order to obtain measurable effects.

The microstructures of the as-sprayed sample, 40 mm/s rapid heat-treated sample, 10 mm/s rapid heat treated sample and the fully annealed sample are shown in Fig. 4. The porosity of the coatings is evident throughout. A higher impact velocity is required to achieve fully dense coatings which would require the use of helium. The etched cross-sections highlight the particle boundaries more clearly, and show the grains within the particles. Despite the change in hardness of the coatings due to the heat treatments, there is no obvious difference between the grain sizes or the overall microstructure of the coatings (i.e. no significant grain growth) due to any of the heat treatment processes based on the images obtained. The change in hardness may be attributed to a reduction in compressive residual stresses, or the precipitation of the alloying elements from solution. Deformation of the particles is evident but not prevalent through the coatings which again highlights the need for higher particle impact velocities. There is also no evidence of mechanical damage such as cracking due to the thermal cycling which occurs during the heat treatment in Fig. 4 b, c, f and g.

The effect of the in-process and traditional heat treatments on the UTS of the coatings is presented in Fig. 5 and Table 2. The as-sprayed samples failed at an average stress of 16 MPa which is considerably lower than that of the bulk material. This is due to the porosity of the coating and the poor interparticle bond strength which is delivered by the coating when sprayed with nitrogen at these processing parameters. However, on average, the UTS is improved for all of the heat treatment procedures carried out. The fully annealed sample delivered the largest improvement with an increase of over 110% in the UTS. The stress relieved sample, which had a much shorter heat treatment time, showed a similar increase in the UTS with over 105% when compared to the as-sprayed state. Both the in-process heat treatments also showed measurable improvements in the UTS of the coatings with a 24% and 52% increase in the UTS for the 40 mm/s and 10 mm/s traverse speeds, respectively. The variation in the UTS also tends to reduce with an increase in the length of the heat treatment time indicating that any defects in the coatings are being repaired or interparticle bond strength is increasing. However, despite the decrease in the variability of the UTS, the variation across the samples is still quite large. This can be attributed to the porosity of the coating. In porous coatings, the failure will be dependent on the presence of large pores or groups of smaller pores which act as stress concentrations causing local yielding. In CS, the pores are not evenly distributed, and hence localised porosity may cause a specimen to fail prematurely. In order to decrease the variation between dog bone specimens from the same coating sample, a lower porosity coating should be manufactured by utilising higher gas velocities. It is also noted that the difference between the UTS delivered by the shorter 1 hour heat treatment is similar to that from the full anneal, which would

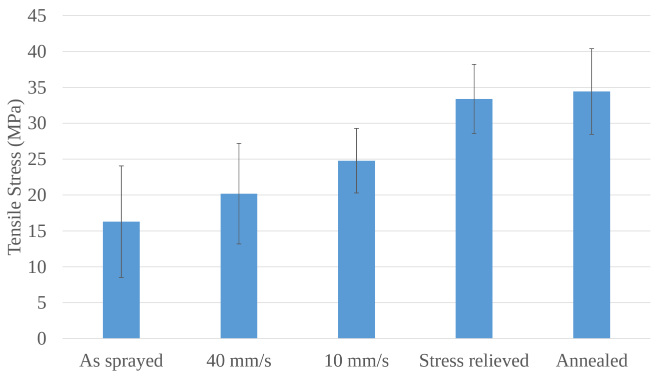


Figure 5: The effect of various heat treatments on the UTS of Al 6061 samples

Figure 6: The effect of various heat treatments on the elongation at failure of Al 6061 samples.

Table 2: Summary of the mechanical properties of the coatings with two standard deviations for 95% confidence intervals

Sample	UTS (MPa)	Strain at failure	Number of tensile samples	Hardness (Hv)	Number of hardness measurements
As Sprayed	16.29 ± 7.77	0.62 ± 0.23	3	60.87 ± 10.60	6
40 mm/s	20.18 ± 7.00	0.56 ± 0.28	4	62.57 ± 11.66	5
10 mm/s	24.78 ± 4.50	0.88 ± 0.32	5	52.23 ± 10.17	5
Stress Relieved	33.39 ± 4.81	0.89 ± 0.16	4	34.25 ± 5.16	5
Annealed	34.42 ± 5.97	1.33 ± 0.49	5	31.17 ± 7.95	5

suggest that longer heat treatments may not be required.

A similar result is shown with the elongation at break of the specimens for various heat treatments as presented in Fig. 6. The annealed sample again shows the greatest improvement with the elongation at break with a 114% increase on average for the 5 samples. The stress relieved sample delivers an improvement in the UTS with a 43% increase, indicating that the ductility of the coating may be more closely linked to the heat treatment time rather than the UTS and hence limited by the significantly shorter duration of the heat treatment. On an average basis, the 10 mm/s in process heat treatment also improved the elongation at break by 43%, but the error bars encompass a significant proportion of this increase. However, the hardness data in Fig. 3 and the UTS data in Fig. 5 would suggest that the 10 mm/s test had a positive effect on the coating's mechanical properties. The 40mm/s test on average has a slightly lower elongation at break when compared to the as-sprayed sample, although the confidence interval overlaps that of the as-sprayed sample. As discussed previously, this heat treatment traverse speed may be too low to provide a conclusive result given the experimental uncertainty and the porosity of the coatings.

Overall, this technique has shown promise in improving the mechanical properties of CS coatings in-process, potentially negating the need for the heat treatment of a component after manufacture. Also, the method described is particularly suitable for the localised heat treatment of large components or components repaired by CS which are not possible to heat treat conventionally. This technique can be carried out with either a laser or a focused infrared device, although the power output of focused infrared devices is ultimately limited to a much greater degree than a laser. However, focused IR technology comes with the advantages of a much lower cost and with fewer safety concerns when compared to a medium to high powered laser system.

Future work

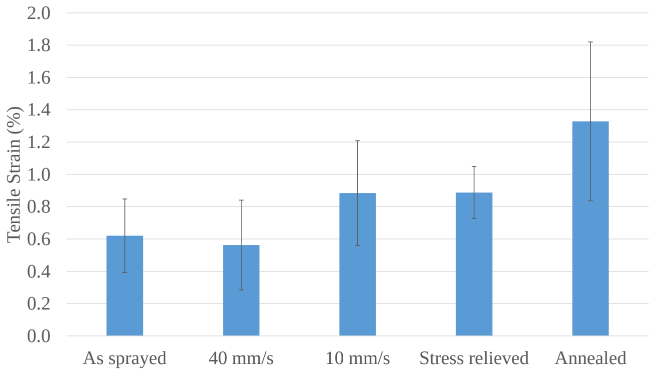
The following work is suggested in order to improve upon these results:

- Employ an argon shield gas to reduce the effect of oxidisation during the heat treatment process. This may potentially improve interparticle bonding and overall performance of the coatings.
- Develop a system to accurately measure the peak temperature reached by the coatings. The surface of the coatings is covered by the focused IR device while processing and therefore measurement using a pyrometer is not possible. Thermocouple embedment is a potential technique which could be investigated.
- Manufacture coatings with a higher density in order to reduce variation in the mechanical properties of the coating. Helium as a processing is suggested in order to provide more deformation and improved mechanical properties.
- Further tests on the traverse speed and the number of passes of the heat treatment should be carried out in order to optimise the process with this focused IR device.
- In order to achieve higher power inputs and faster processing speeds, a laser may be used at a higher initial cost than focused infrared heating technology.

Conclusions

A focused infrared device has been employed to rapidly heat treat CS coatings in process with the aim of improving the mechanical properties of the coatings. The following conclusions can be drawn from this work:

- The rapid heat treatment showed improvements in the mechanical properties of the coating for the two traverse speeds tested with a 24% and 52% increase in the UTS of the coating for the 40 mm/s traverse speed and the 10 mm/s traverse speed, respectively. This was compared to a fully annealed sample which showed the greatest improvement in the UTS with a 110% improvement.
- The elongation at break also showed improvements, for the slowest speed tested, although the uncertainty associated with this result was relatively large.



- The hardness of the coatings was reduced, reinforcing the hypothesis that the rapid heat treatments positively affected the mechanical properties of the coatings.
- This work demonstrates that the rapid in process heat treatment of coat spray coatings is possible and that the heat treatment examined delivers improvements in the mechanical properties of Al 6061 coatings.

Acknowledgments

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