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SPRAY-FORMING MONOLITHIC ALUMINUM ALLOY AND METAL MATRIX COMPOSITE STRIP

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ABSTRACT

Spray forming with de Laval nozzles is an advanced materials processing technology that converts a bulk liquid metal to a near-net-shape solid by depositing atomized droplets onto a suitably shaped substrate. Using this approach, aluminum alloys have been spray formed as strip, with technoeconomic advantages over conventional hot mill processing and continuous casting. The spray-formed strip had a flat profile, minimal porosity, high yield, and refined microstructure. In an adaptation to the technique, 6061 Al/SiC particulate-reinforced metal matrix composite strip was produced by codeposition of the phases.

SPRAY FORMING is an advanced materials processing technology that combines rapid solidification processing (RSP) with product shape control. The interaction of a high velocity gas jet with a liquid metal stream or sheet atomizes the metal, producing a spray of fine droplets that are deposited onto a substrate or pattern to form a solid. Spray forming with de Laval (converging/diverging) nozzles was developed at the Idaho National Engineering Laboratory (INEL) to process metals, polymers, and metal matrix composites (MMCs) from a bulk liquid to net shape or near-net shape in a single step [1-7]. In this approach, a liquid is aspirated or pressure-fed into a de Laval nozzle. There it contacts a high-velocity, high-temperature inert gas that disintegrates the liquid into very small (~20 μm) droplets and entrains the droplets in a highly directed spray.

Spray deposition with de Laval nozzles typically involves *transonic* gas-particle flow through the nozzle and subsonic free jet flow from the nozzle to the substrate [8]. Laser Doppler velocimetry [9] has established that droplets are accelerated in the flow field to velocities of about 50 m/s. After exiting the nozzle, the spray jet rapidly entrains large volumes of relatively cold inert gas, which removes the liquid metal's superheat and approximately 75% of the enthalpy of solidification. As a result, droplets arrive at the substrate in semi-solid, solid, and undercooled states depending on nozzle design, operating parameters, and droplet size and trajectory in the flow field. Upon impacting the substrate, the droplets weld together, replicating the shape and surface texture of the substrate or pattern, while releasing the remaining enthalpy by convection and conduction through the substrate.

Using this approach, aluminum alloys have been spray formed as strip. The spray-formed strip had a flat profile, minimal porosity, high yield and refined microstructure. 6061 Al/SiC particulate-reinforced MMC strip was also produced by codeposition of the phases.

EXPERIMENTAL

The apparatus used to produce monolithic and composite aluminum strip has been described elsewhere [8]. The alloy to be sprayed is induction melted under a nitrogen atmosphere, superheated about 150°C, and pressure-fed into a de Laval spray nozzle of our own design. Nitrogen gas is used to atomize the metal, entrain the droplets, and deposit them onto a grit-blasted steel drum. Droplets impact the drum, positioned about 0.3 m (12 in.) from the nozzle, producing a strip of metal 2.5-13 mm (0.1-0.5 in.) thick, depending on conditions. An inert gas atmosphere within the spray apparatus minimizes slag formation in the melt and in-flight oxidation of the atomized droplets.

The nozzle is operated at a static pressure, measured at its inlet, of about 172 kPa absolute (25 psia). The temperature of the atomizing gas has been varied from 20 to 800°C with acceptable results. A gas-to-metal mass flow ratio (G/M) of about 0.3 is typically used, and metal mass flow rates are in the range 8,900 to 54,000 kg/h per meter (500 to 3000 lb/h per inch) of nozzle width transverse to the flow direction. This nozzle dimension is scaled for the desired strip width. To date, most experiments have been conducted using bench-scale (17 mm wide) nozzles.

Composite strip was produced by codepositing the phases onto a water-cooled, grit-blasted, mild steel drum. The ceramic phase was pressure-fed into the nozzle in the form of an aerosol upstream of the molten metal, which was also pressure-fed into the nozzle. Heat transfer from the atomizing gas was used to adjust the temperature of the ceramic particulate as it accelerated through the nozzle to the liquid metal atomization region. This approach provided independent control of the feed rates of the liquid metal and ceramic, good component mixing, and independent control of the temperatures of the metal and ceramic inside the nozzle.

During a typical MMC run, 6061 aluminum alloy was induction heated to about 150°C above the liquidus temperature and atomized with argon heated to about 750°C. Aluminum throughput was as high as 29,000 kg/h-m (0.8 ton/h-in.), with a corresponding gas-to-metal mass flow ratio (G/M) of 0.1. G/M values as high as 7 were found to give acceptable results.

RESULTS AND DISCUSSION

Monolithic Strip. The transverse cross section of 6 mm (0.25 in.) thick aluminum strip shown in Figure 1 was spray formed with a bench-scale nozzle. A flat profile is critical for this application to prevent fracture during subsequent rolling operations. Overspray losses, defined as unconsolidated particulate and thin edge trimmings, are about 9% for bench-scale nozzles. Preliminary results indicate that overspray decreases as the nozzle is scaled-up.

In the spray jet, airborne droplets cool by convection and radiation. The relative contributions of both cooling mechanisms depend on droplet temperature, Weber number, gas and droplet thermal diffusivity, and other factors. Under most conditions, convection cooling strongly dominates. The cooling rate of droplets in 6061 aluminum spray jets was estimated by measuring the dendrite cell size in polished/etched powders (see Figure 2). Powder was partitioned into size bands using sieves of 300, 212, 177, 149, 125, 75, 63, 45, 38, 25, 20, 15, 10, and 5 µm. In general, dendrite cell size increased with increasing powder size, consistent with previously published results on gas atomized aluminum alloys [10-14]. For example, the cell size increased from about 1.8 µm for a 20 µm particle, to about 9 µm for a 200 µm particle. Cell size was found to follow a power law relationship with powder size. Cooling rate was estimated from measured dendrite cell size using the relationship

$$\lambda=B\epsilon^{-n}$$

where λ is the average dendrite cell size, ϵ is the cooling rate, and B and n are material and process dependent constants [10-14]. For aluminum alloys, they are typically about 50 $\mu m(Ks^{-1})^n$ and 1/3, respectively, over the range 10^{-5} to 10^6 K/s [10]. Cooling rates varied inversely with droplet size, ranging from about 10^2 to 10^4 K/s, placing them well within the range of rapid solidification and at least one order of magnitude

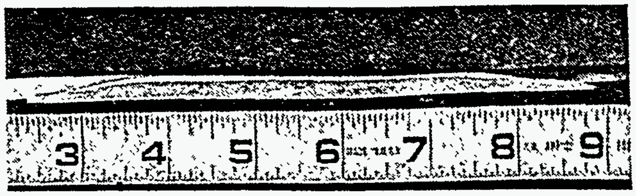
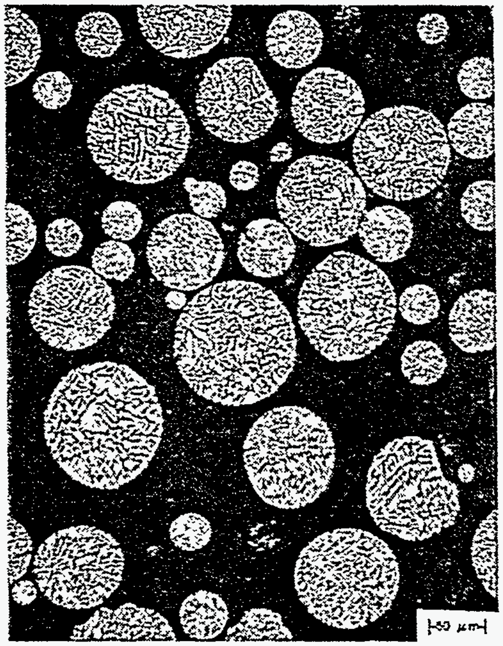
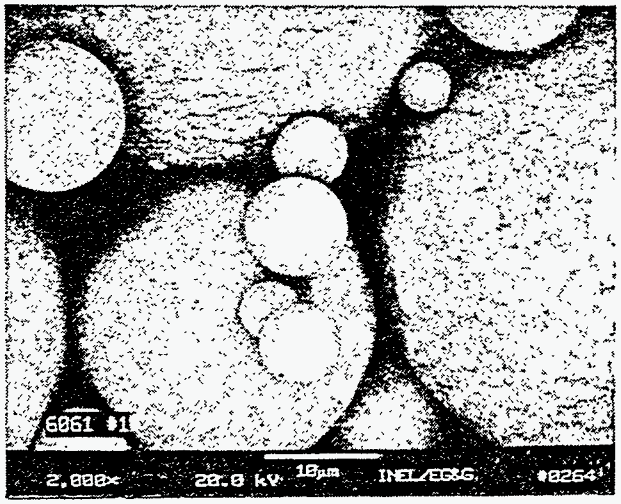


Fig. 1 - Transverse cross section of 6061 aluminum alloy strip spray formed using a bench-scale de Laval nozzle.



(a) Photomicrograph of polished/etched powder showing fine-scale dendrite/cellular structure.



(b) SEM photograph showing surface morphology of powder.

Fig. 2 - 6061 aluminum overspray powders.

higher than the average cooling rate of the bulk deposit (Figure 3).

Discrete droplet impacts at the exposed deposit surface of 3003 aluminum strip, shown in Figure 4, provide insight into the mechanism of equiaxed grain formation. Dendrites within the solidified particles are clearly visible in both photographs. Dendrite cells fragment after impact to provide a high concentration of nuclei, which help refine the microstructure. The degree of droplet spreading in Figure 4a suggests a high liquid fraction. Dendrite debris, fine-scale entrapped gas, and a high cooling rate at the surface may help explain why coarsening is inhibited in spray-formed materials. The early stages of grain growth and coarsening are observed within two regions of the splat before the prior splat boundary has been erased. Equiaxed grain formation is at an advanced stage of development within one or two droplet diameters from the surface of the deposit. The droplet in Figure 4b exhibited low shear at impact, suggesting a low liquid fraction. Some dendrite fragmentation and alignment is observed within the core of the droplet.

Metallography of as-deposited 3003 aluminum indicates a refined equiaxed microstructure with good, constituent dispersion and no macrosegregation. Depending upon conditions, average grain size is 15 to 50 µm for this alloy. The photomicrograph in Figure 5a was taken near the center of a strip. Hot rolling at 450°C to 44% thickness reduction was found to reduce the average grain size by 25% (Figure 5b). The as-deposited bulk density, measured by water displacement using Archimedes' principle, is 95 to 99.5% of theoretical density. Porosity in the samples is largely "cold" porosity and tends to be concentrated at the deposit/substrate interface. "Cold" porosity is formed when the liquid fraction of impinging droplets is insufficient to fill pores in the strip due to rapid quenching and rapid droplet arrest conditions. These conditions are favored at the substrate. "Hot" porosity, on the other hand, is characterized by circular pores formed by gas engulfment during solidification. Hot porosity normally was not observed. Efforts to reduce porosity in the strip near the deposit/substrate interface have been successful; the as-deposited material shown in Figure 5c is an example.

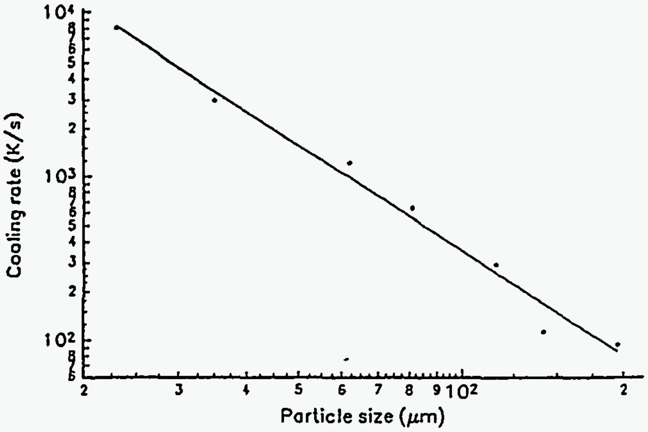
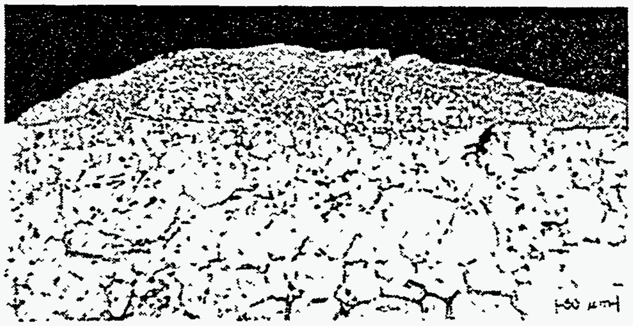


Fig. 3 - Cooling rate of 6061 aluminum droplets.

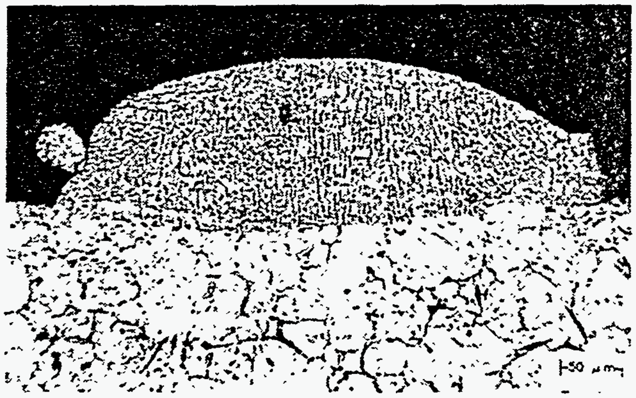
Preliminary room temperature tensile properties were determined for spray-formed 3003 aluminum after tempering to conditions commonly available for commercial strip. As-deposited samples (without scalping or machining) were hot rolled to 50% thickness reduction in a single pass, annealed, and cold rolled in one or two passes to yield -H14, -H16, and -H18 tempers. Results summarized in Table I compare commercial 3003 aluminum strip with these tempers [15] with the as-deposited, unprocessed strip, and as-deposited strip (without scalping or machining) cold rolled to 50% thickness reduction. Ranges in values for spray formed material reflect differences in experimental conditions.

Particulate-Reinforced MMC Strip. As-deposited composite strip was sectioned, heated to 450°C in an argon-purged furnace, and hot rolled to 80% thickness reduction followed by quenching. Samples were then solution heat treated and precipitation hardened to yield a -T6 temper. Depending on spray conditions, particulate volume fraction ranged from 4 to 15%, as determined by acid dissolution of the matrix. Optical microscopy of polished samples indicated a uniform distribution of particulate in the matrix phase; an example is given in Figure 6.

Room temperature tensile properties were determined for spray-formed and hot-rolled matrix and 4 vol% composite

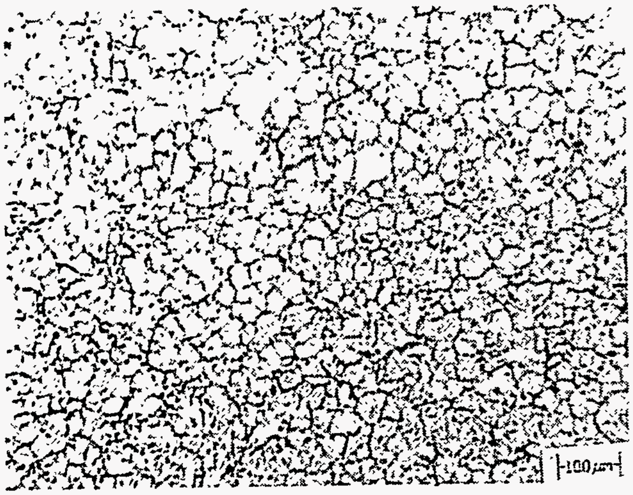


(a) High liquid fraction droplet.

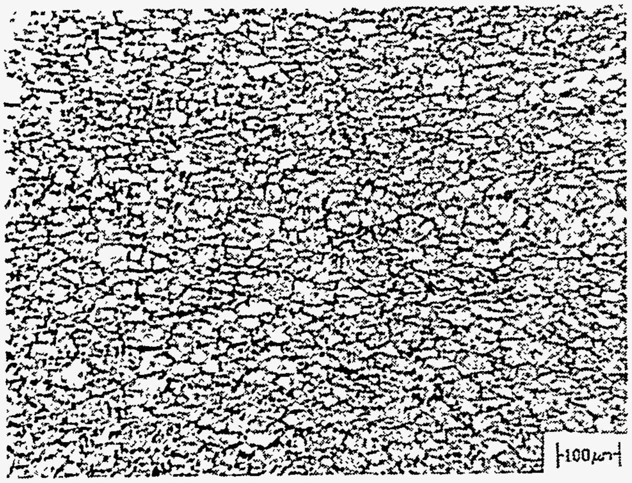


(b) Low liquid fraction droplet.

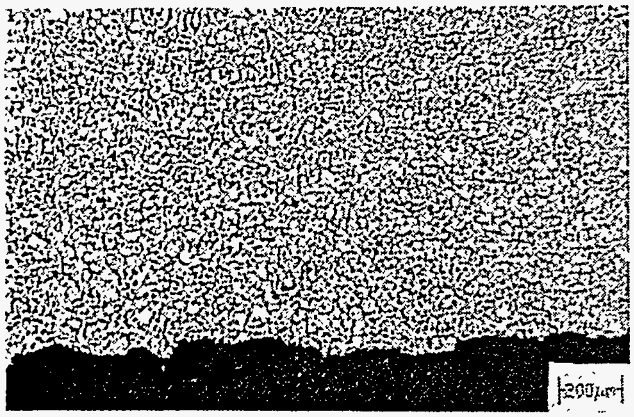
Fig. 4 - Photomicrographs of discrete droplet impacts at exposed deposit surface of spray-formed 3003 aluminum alloy strip.



(a) As-deposited bulk microstructure, 28 µm average grain size.



(b) Bulk microstructure after hot rolling at 450°C to 44% thickness reduction. 21 µm average grain size.



(c) Grain structure of as-deposited material at deposit/substrate interface, 25 µm average grain size.

Fig. 5 - Microstructure of spray-formed 3003 aluminum. Keller's Etch.

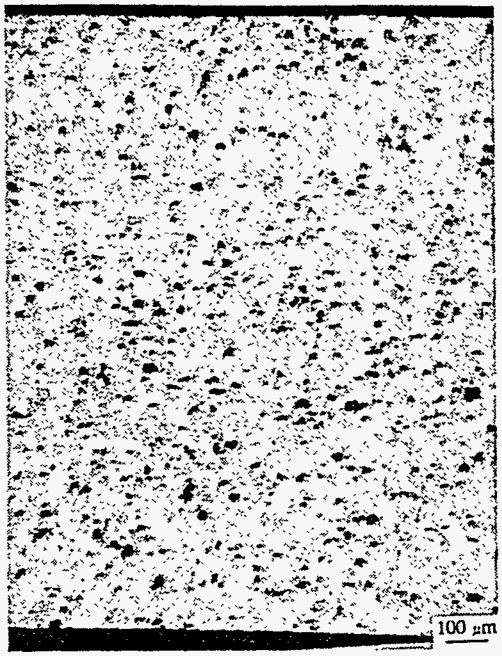


Fig. 6 - Photomicrograph of 15 vol.% 6061/SiC composite strip hot rolled to 80% thickness reduction. Polished, unetched.

samples; these results are summarized in Table II. Both materials showed improvements (about 10%) in ultimate strength and yield strength over commercial 6061-T6 strip, but a reduction in elongation. The unreinforced spray-formed and hot-rolled material also had an elastic modulus about 10% higher than that of commercial 6061-T6. The composite material exhibited a notable increase in modulus (about 33%). These results are very encouraging but should be viewed as preliminary. Evaluation of a larger number of samples is necessary to optimize spray conditions and to establish statistical validity.

CONCLUSIONS

1. De Laval nozzle spray forming can produce aluminum strip at high production rates. Technoeconomic analysis indicates this spray-forming approach is competitive with continuous casting and hot mill processing of high volume sheet alloys.

2. Spray-formed aluminum alloy strip has a flat profile, low porosity, and high yields. Refined, equiaxed grain structures and uniform distribution of fine constituent particles and dispersoids are observed. Low porosity in the deposit at the deposit/substrate interface eliminates the need for scalping. Total elimination of porosity has not yet been achieved using a room temperature substrate. Tensile properties of spray-formed strip compare favorably with those of commercial strip for commodity aluminum alloys tested (3003 and 6061).

Table I. Tensile Properties[a] of Commercial 3003 Aluminum Alloy and Spray-Formed Strip

Material/Temper or Condition	Ultimate Strength, MPa (ksi)	Yield Strength, MPa (ksi)	Elongation, %[b]
Commercial/ -O	97 (14.0)	34 (5.0)	25
Commercial/ -H14	138 (20.0)	117 (17.0)	5
Commercial/ -H16	165 (24.0)	145 (21.0)	3-4
Commercial/ -H18	186 (27.0)	165 (24.0)	2
Spray formed/ -H14	160.0-173.1 (23.2-25.1)	154.4-170.3 (22.4-24.7)	3.4-7.8
Spray formed/ -H16	195.5-198.6 (28.8-29.7)	188.2-195.5 (27.3-28.5)	3.0-6.8
Spray formed/ -H18	228.9 (33.2)	207.5 (30.1)	2.7
Spray formed/ as-deposited	129.3-135.1 (18.8-19.6)	86.2-102.2 (12.5-14.8)	9.5-14.6
Spray formed/ cold rolled 50%	190.8-194.4 (27.7-28.2)	186.2-188.1 (27.0-27.3)	5.0-5.2

a. Values for commercial sheet are minimum specification limits for sheet of comparable thickness to spray-formed sheet with the same temper.

b. For commercial material, % elongation is in 50 mm (2 in.) gage length. % elongation for spray formed samples is in 25 mm (1 in.) gage length.

Table II. Tensile Properties of 6061 Al and 6061 Al/SiC Strip.

Sample	Yield Strength, 0.2% Offset, MPa (ksi)	Ultimate Strength, MPa (ksi)	Elongation in 50 mm, %	Elastic Modulus, GPa
Commercial 6061-T6	277.2 (40.2)	307.5 (44.6)	12.1	70
Spray Formed and Hot Rolled[a] 6061-T6	306.1 (44.4)	320.6 (46.5)	7.4	77
Spray Formed and Hot Rolled[a] 6061-T6/SiC (4 vol %)	307.5 (44.6)	336.5 (48.8)	5.4	93

a. 80% thickness reduction

3. The process can readily be adapted to spray form particulate-reinforced metal matrix composites by codeposition of the phases. Preliminary results with 6061-T6/SiC strip indicates some improvement in strength, a reduction in ductility, and a notable improvement in elastic modulus compared with the commercial matrix strip with the same temper.

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