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Microstructure Evolution During Spray Rolling and Heat Treatment of 2124 Al

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Abstract

Spray rolling is a strip-casting technology that combines elements of spray forming and twin-roll casting. It consists of atomizing molten metal with a high velocity inert gas, quenching the resultant droplets in flight, and directing the spray between mill rolls. In-flight convection heat transfer from atomized droplets and conduction heat transfer at the rolls rapidly remove the metal's latent heat. Hot deformation of the semi-solid material in the rolls results in fully consolidated, rapidly-solidified product. Spray rolling operates at a higher solidification rate than conventional twin-roll casting and is able to process a broader range of alloys at high production rates.

A laboratory-scale strip caster was constructed and used to evaluate the interplay of processing parameters and strip quality for strips up to 200 mm wide and 1.6–6.4 mm thick. Plans are underway to scale to 600 mm width and demonstrate steady-state operation. This paper describes microstructure evolution during spray rolling. It also examines how the liquid content of the semi-solid material, and post-deposition thermo-mechanical processing, influence the microstructure and mechanical properties of spray-rolled 2124 Al.

Introduction

Direct strip-casting technologies are attractive because they improve the economics of manufacturing aluminum sheet products by eliminating the ingot casting, homogenization, and hot-rolling unit operations in conventional ingot processing while significantly reducing energy consumption. Spray rolling is a relatively new strip-casting technology, being developed, that combines elements of spray forming and twin-roll casting [McH04, McH05]. In spray rolling, molten metal is atomized with a high velocity inert gas, the resultant droplets are quenched in flight, and the spray deposits onto mill rolls (**Figure 1**). In-flight convection heat transfer and conduction heat transfer at the rolls rapidly removes the latent heat. Hot deformation of the semi-solid material by the rolls fully consolidates the rapidly-solidified product. The high solidification rate in spray rolling allows a wider range of alloys to be processed, and at higher production rates, than is currently possible in commercial twin-roll casting practices [Gra05, Nai06, Yun00].

The main differences between twin-roll and spray-roll strip casting are:

1. In twin-roll casting, the metal's latent heat is removed almost exclusively by conduction heat transfer to water-cooled rolls. In spray rolling, convection heat transfer from small atomized droplets combines with conduction transfer at the rolls to increase the production rate and limit segregation and casting flaws such as surface bleeds.
2. The metal introduced to the rolls in twin-roll casting is molten, while in spray rolling it is semi-solid. Solid particles in the semi-solid material are uniformly distributed and act as nucleation sites and heat sinks for the remaining liquid.

The interplay of processing parameters and strip quality was investigated using a laboratory-scale strip caster that can produce strips up to 200 mm wide and 1.6–6.4 mm thick. Planned work will produce strips up to 600 mm wide and demonstrate steady-state operation. This paper examines microstructure evolution during spray rolling and explores how gas-to-metal mass flow ratio influences the microstructure and mechanical properties of spray-rolled 2124 Al. The influences of solution heat treatment and cold rolling on grain structure and constituent particle spheroidization are also examined.

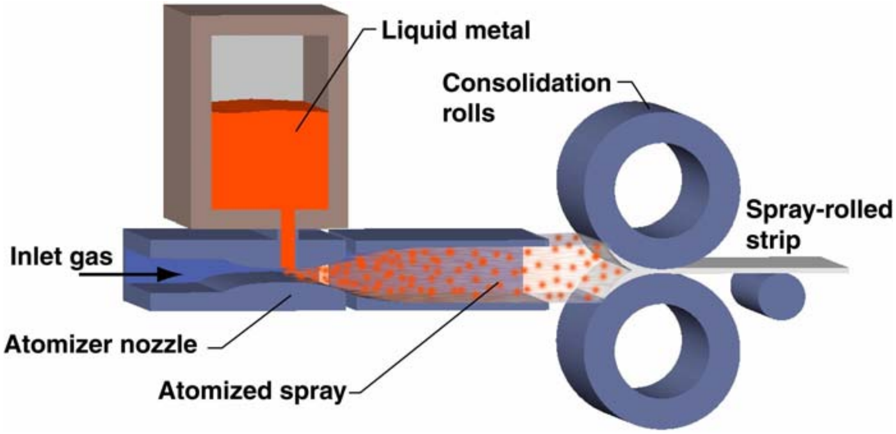

Fig. 1: Schematic of spray rolling approach.

Experimental procedure

The spray-rolling strip caster was designed and constructed at Idaho National Laboratory. 2124 alloy was induction heated under a nitrogen atmosphere, superheated about 100°C above the liquidus temperature, and pressure fed into an atomizer designed and constructed in house. Atomized droplets were directed horizontally and deposited into the roll gap of a 200 mm × 300 mm 2-HI Fenn rolling mill (Model 4-081). The mill had standard tool-steel rolls that were not water cooled. Graphite lubricant was applied to the roll surface prior to a run but was not reapplied during the run. A nitrogen atmosphere within the spray apparatus minimized in-flight oxidation of the atomized droplets. Currently, the caster is not equipped with a coiler; individual strips measuring about 100 mm wide, 4.3 mm thick, and 2 m long were produced by starting/stopping the spray (**Figure 2**). For this study, strips were made using gas-to-metal mass flow ratios (G/M) of 0.15 and 0.3 while maintaining other processing conditions constant. The production rate was 4100 kg/h·m.

Microstructure was evaluated optically with an Olympus Model PME-3 metallograph. Keller's etchant was used to reveal grain structure and highlight constituent phases. X-ray diffraction was performed using Cu (Kα1 + Kα2) radiation on a Bruker Model D-8 Advance system operating at a sampling rate 0.6° (2 theta) per minute. Resulting patterns were analyzed with EVA software.

Tensile testing was performed with an Instron 4505 screw-driven test machine following the ASTM E-8 procedure. Spray-rolled 2124 strips were heat treated to the T851 temper, with cold rolling substituted for the normal commercial practice of stretching. Samples were solution treated at 493°C for 1 h, water quenched, cold rolled 3%, and aged at 190°C for 12 h. Additional samples were soaked at 493°C for 2 h, water quenched, cold rolled to a 0-75% thickness reduction, and annealed at 493°C for 10 min.

Fig. 2: Typical spray-rolled and edge-trimmed 2124 Al strip used in this study.

Results and discussion

Microstructure evolution during spray rolling

In spray rolling, the rolls perform several functions. They collect spray, extract remaining latent heat, and consolidate semi-solid material to form fully-dense strip. By the time the spray impacts the rolls, most of the metal's latent heat has been removed by convection heat transfer to the relatively cold gas that is entrained into the jet. The remaining liquid phase that deposits out has the same composition as the liquid metal feed. This greatly limits segregation because the surrounding solid material acts as a heat sink to rapidly solidify the remaining liquid phase. Thus, unlike twin-roll casting, solidification does not initiate at the wall of the rolls and progress inwardly. Nevertheless, the last material that solidifies can be enriched in alloying additions. When processing alloys with a broad freezing range such as AA2124 (**Table 1**), it is important to limit the volume of liquid in the semi-solid material to control casting flaws such as surface bleeds and sub-surface stringers of constituent phases. Experience has shown that this is most easily and effectively accomplished during droplet formation and cooling rather than post-deposition thermo-mechanical processing (TMP).

Table 1: Nominal composition and melting range of 2124 aluminum alloy

Alloy	Composition	Melting Range
2124	Al-4.4 Cu-1.5 Mg-0.6 Mn	502–638°C

To examine microstructure evolution during spray rolling, the rolls and spray were stopped simultaneously during operation of the caster. An illustration of the resulting wedge is given in **Figure 3.** A cross section of the wedge revealed four distinct regions:

1. Impact region where the spray is collected by the rolls
2. Consolidation region where the semi-solid material is funneled and is initially compacted
3. Hot rolling region near the roll nip
4. Product strip downstream of the nip.

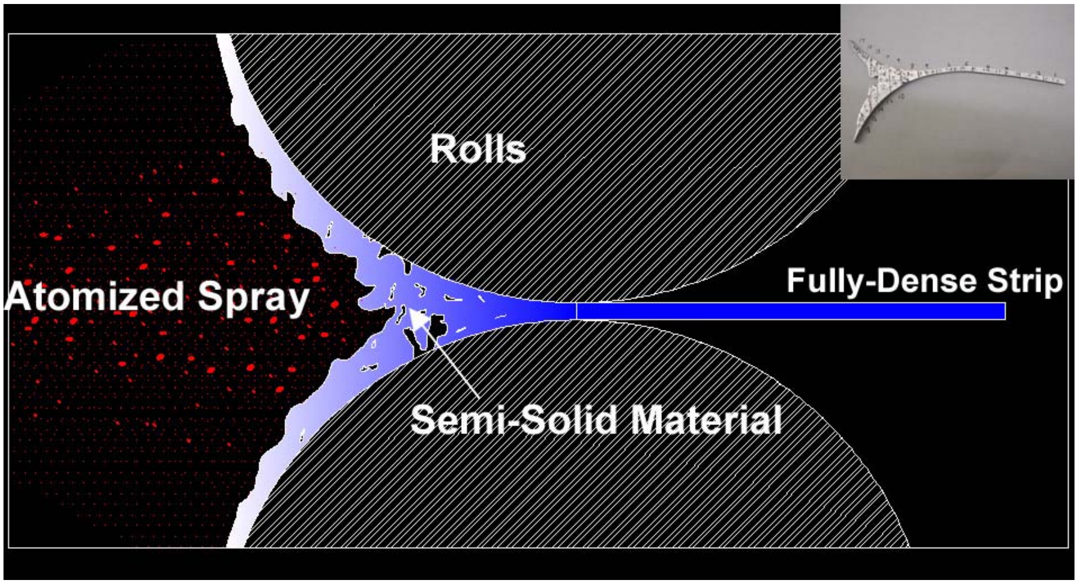

Fig. 3: Depiction of the spray rolling process showing collection and consolidation of the atomized spray by counter-rotating rolls to form fully-dense strip. Inset figure shows AA2124 "wedge" formed by simultaneously stopping spray and rolls.

Figure 4 illustrates representative microstructures in each of these regions. The impact zone is characterized by an equiaxed, fine grain structure with random, isolated pores of various shapes. Grains at the roll surface were slightly smaller in size than interior grains but remained equiaxed. This region most closely resembled what is typically observed when spraying onto a flat surface.

The consolidation region also consisted of grains with an equiaxed structure but contained a decreasing concentration of pores as the material progressed toward the nip. Grains near the surface were elongated in the rolling direction. The high curvature of the small (200 mm diameter) rolls used in this work and high deposition rate resulted in rapid funneling of the semi-solid material, and many macroscopic flaws were observed in this region, such as folds and tears, as depicted in **Figure 3**. These flaws mostly originated at the roll surface and were likely due to strip detachment from the rolls and back slip as the pressure gradient increased.

In the densification region, porosity was eliminated at a point where the cross-sectional thickness was about twice the roll gap. Grain morphology transitioned from equiaxed to somewhat elongated upstream of the roll gap, suggesting the material was completely solidified at that point and experienced some warm rolling.

Thermo-mechanical processing of AA2124

When processed by spray rolling, the microstructure and properties of 2124 product strip are sensitive to the solid fraction of the spray when it impacts the rolls. Solid fraction can be controlled by adjusting G/M, which influences average droplet size in the spray and in-flight droplet cooling rate. Strip samples produced at G/M values of 0.15 and 0.30 showed differences in the distribution of the solute-rich phase and resultant tensile properties in both the as-spray-rolled and T851 temper states. **Figure 5** compares the microstructure of ingot-cast 2124 with as-spray-rolled strips produced at two G/M values. The ingot-cast material is characterized by a

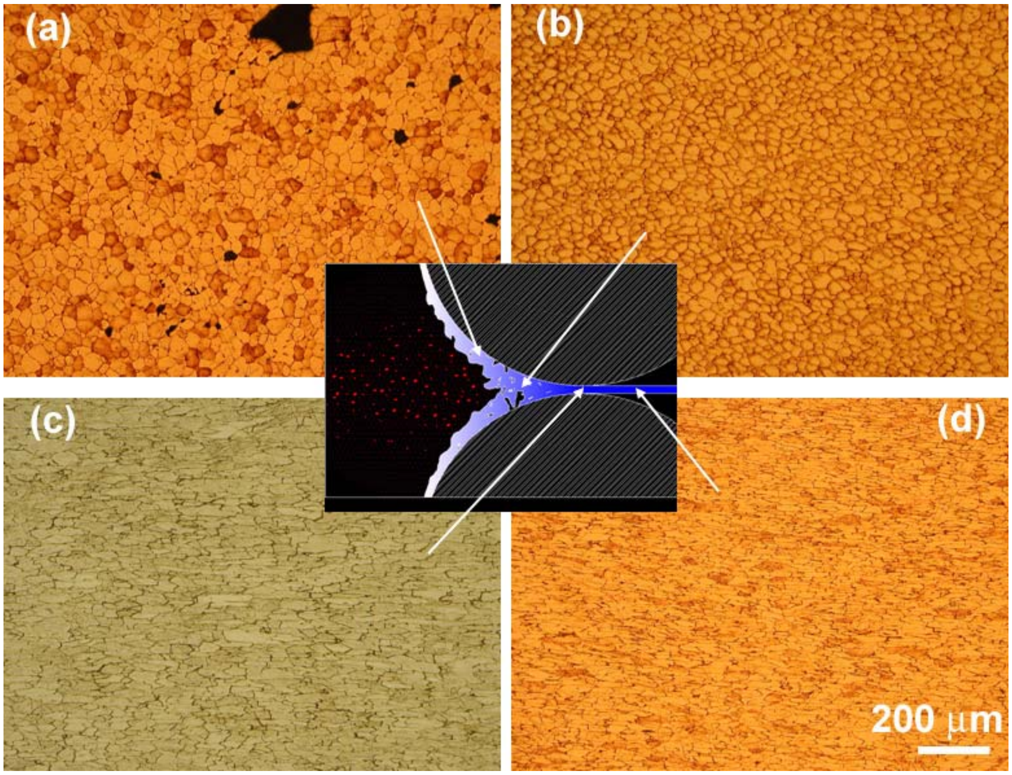

Fig. 4: Longitudinal photomicrographs illustrating microstructure evolution of AA2124 during spray rolling at G/M=0.15. (a) Spray collection region. (b) Initial compaction of semi-solid material. (c) Densification near roll nip. (d) Fully-dense strip exiting the rolls.

coarse dendritic structure and extensive interdendritic segregation of the solute-rich phases. In contrast, strip-cast material exhibited a fine-grain size (about 15 μm) and nearly equiaxed structure with relatively small, angular precipitates of Al-Cu-Mg-Mn phase around grain boundaries. Closer analysis of grain structure using TEM revealed a subgrain structure and an average grain size of 1 to 2 μm (**Figure 6**).

The amount and distribution of Al-Cu-Mg-Mn precipitates can be varied by controlling G/M. **Figure 7** compares the microstructure viewed in the longitudinal (rolling) direction of as-spray-rolled strip processed at G/M = 0.15 and 0.30 near each rolling surface and middle of the strip. Higher concentrations of undissolved Al-Cu-Mg-Mn phase are observed for the strip spray rolled at G/M = 0.15, particularly near the surfaces of the strip. This is due to the solute-rich phase being squeezed toward the surface during compaction in the roll gap. Tempering spray-rolled 2124 to the T851 condition resulted in a recrystallized grain structure with spheroidization of constituents (**Figure 8**). The microstructures of longitudinal sections of strip samples produced at G/M = 0.15 and 0.30 were similar in terms of grain size and morphology (**Figure 8a and 8b**), with little grain elongation compared to commercial 2124 (**Figure 8c**) due to the relatively modest amount of rolling during processing. A 2-h solution heat treatment at 493°C did not completely redistribute the solute-rich phases in material processed at 0.15 G/M. Particles were observed to spheroidize and diffusion processes resulted in clusters of discrete particles of Al_2Cu, and Al_2CuMg, with smaller amounts of Mg_2Si, (Fe,Mn)Al_6, Cu_2FeAl_7 , Cu_2MnAl_{20}, etc.

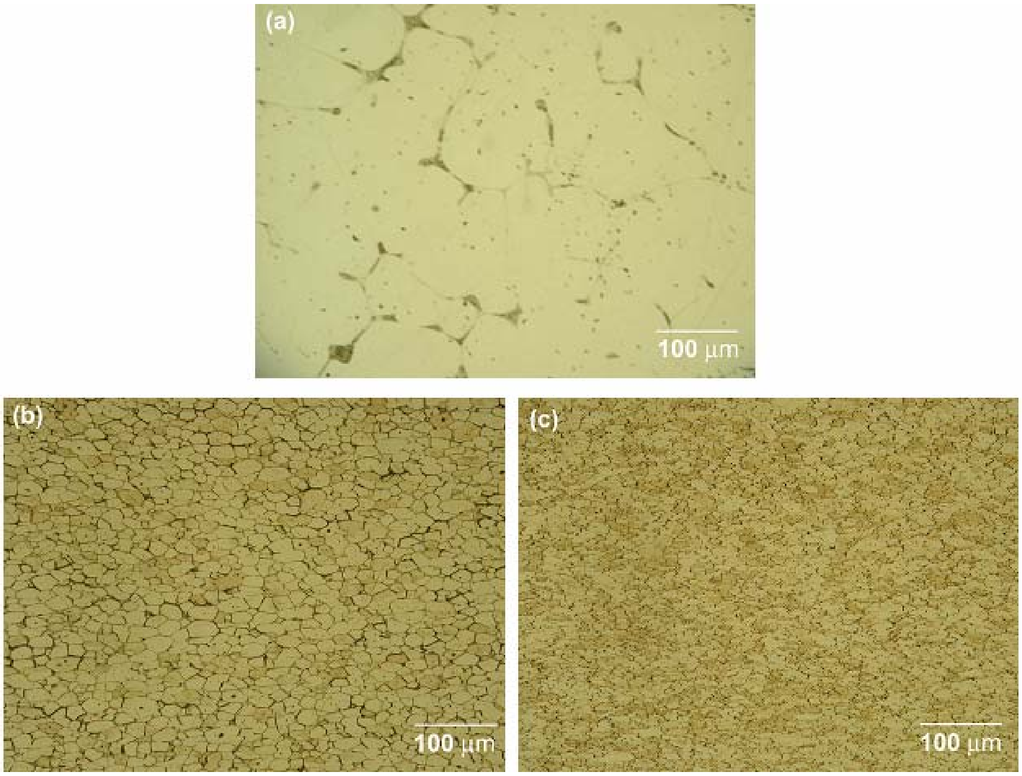

Fig. 5: Transverse photomicrographs of 2124 aluminum. (a) Ingot cast with slow solidification. (b) As-spray-rolled using G/M = 0.15. (c) As-spray-rolled using G/M = 0.30.

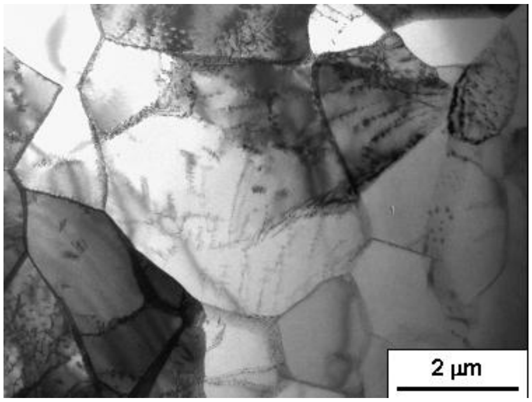

Fig. 6: TEM photomicrograph of 2124 Al as-spray-rolled at G/M=0.15.

as shown in **Figure 9**. X-ray diffraction analysis of as-spray-rolled 2124, spray-rolled 2124-T851 and commercial 2124-T851 is summarized in **Figure 10**. Scan profiles of both 2124-T851 strips were similar. Primary Al peaks in the as-spray-rolled 2124 scan were shifted somewhat due to lattice strain, and show more clearly resolved peaks corresponding to Al_2CuMg and Al_2Cu constituent phases.

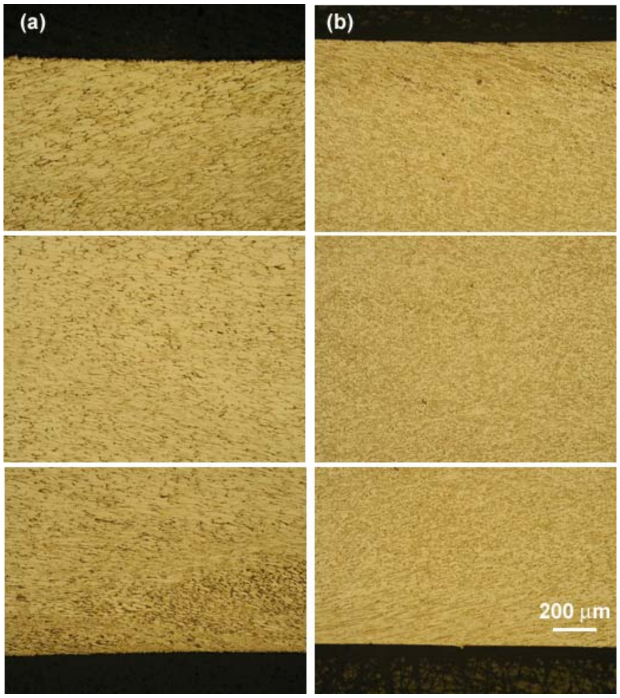

Fig. 7: Longitudinal photomicrographs of as-spray-rolled 2124. (a) Spray rolled at G/M = 0.15 near each rolling surface (top, bottom) and near the center of a 4.3 mm thick strip. (b) Same as (a) but spray rolled at G/M = 0.30.

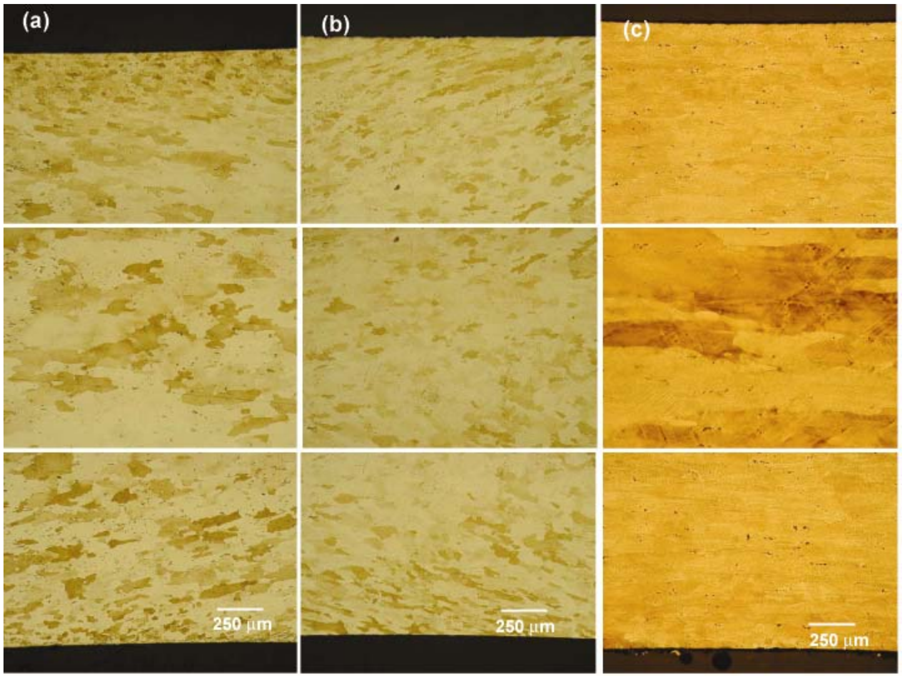

Fig. 8: Longitudinal photomicrographs of 2124-T851 aluminum. (a) Spray rolled at G/M = 0.15 near each rolling surface (top, bottom) and near the center of a 4.3 mm thick strip. (b) Same as (a) but spray rolled at G/M = 0.30. (c) Commercial plate near each rolling surface (top, bottom) and near the center.

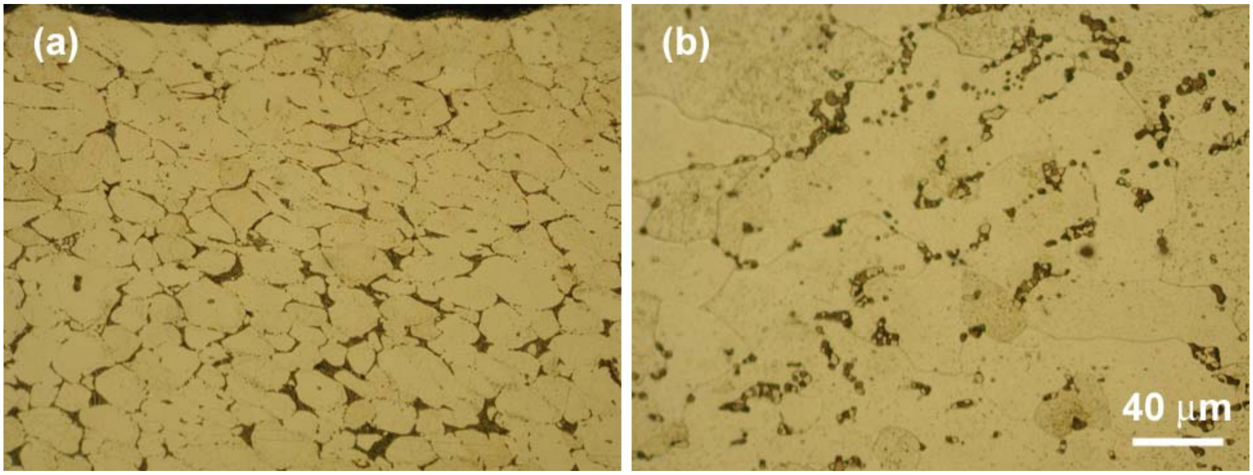

Fig. 9: Photomicrographs illustrating micro-segregation in as-spray-rolled 2124 (G/M=0.15) and redistribution of the solute-rich phase following solution heat treatment. (a) As-spray-rolled at G/M = 0.15 with angular Al-Cu-Mg-Mn precipitates near grain boundaries. (b) Diffusion and spheroidization resulting in clusters of discrete Al_2Cu (light) and Al_2CuMg (dark) constituent particles following a 2-h solution heat treatment.

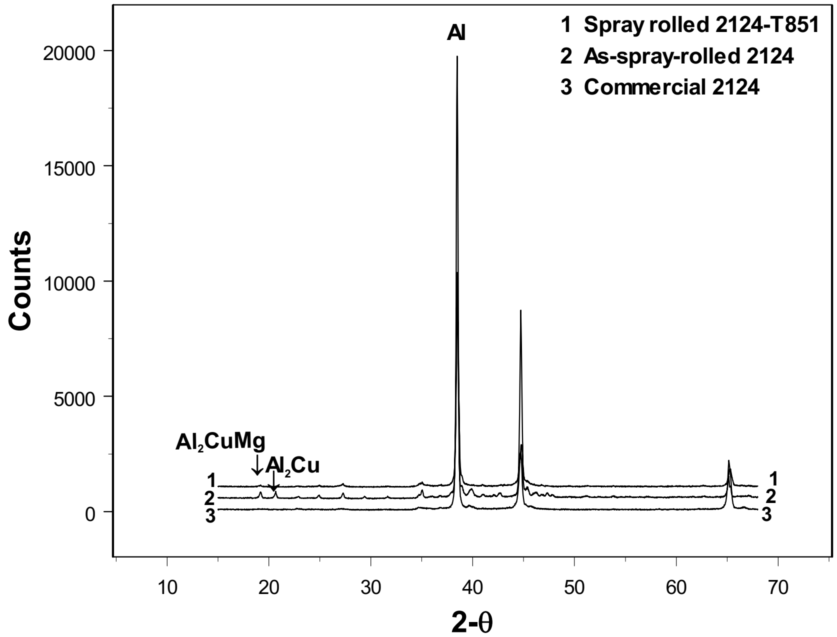

Fig. 10: X-ray diffraction scans of as-spray-rolled 2124, spray-rolled 2124-T851, and commercial 2124-T851.

Tensile properties of as-spray-rolled 2124 strip and 2124-T851 strip at G/M=0.15 and 0.30 are summarized in **Table 2**. Results indicate that increasing the solid fraction of semi-solid 2124 introduced into the roll gap improves tensile properties. For as-spray-rolled 2124 strip, increasing

G/M from 0.15 to 0.30 resulted in an increase in ultimate tensile strength (UTS) of 39%, an increase in yield strength (YS) of 46%, and a doubling of the ductility. This is largely due to a reduction in micro-segregation, particularly at the strip surface, and more uniform distribution of constituent phases at G/M = 0.30. This trend remained following tempering to the T851 condition. Increasing G/M from 0.15 to 0.30 resulted in increases in UTS, YS, and % elongation of 9%, 15%, and 17%, respectively. As shown in **Table 2**, the tensile properties of 2124-T851 produced at G/M = 0.30 compare favorably with those of commercial material. Tensile properties of transverse and longitudinal sections cut from spray-rolled strip were very nearly identical, in contrast to commercial material which was more sensitive to rolling direction.

Table 2: Tensile properties of as-spray-rolled 2124 and 2124-T851.

Condition	G/M	Ultimate Tensile Strength (MPa)	Yield Strength (MPa)	Elongation at Failure (%)
Commercial-T851	—	483	448	6
As spray rolled	0.15	228	179	5
As spray rolled	0.30	317	262	10
Spray rolled –T851	0.15	441	407	6
Spray rolled –T851	0.30	483	469	7

The grain morphology of a recrystallized alloy depends on many factors, such as the amount of cold work, recrystallization time and temperature, and solidification rate during processing. Secondary-phase particles play an important role in recrystallization behavior. Recrystallized grain morphology of spray-rolled 2124 (G/M = 0.15) was examined by cold-rolling solution heat treated samples up to 75% followed by a 10 min anneal at 493°C. Edge cracking during cold rolling was minimal. Samples were sectioned in the longitudinal (**Figure 11**), short transverse (**Figure 12**), and long transverse (**Figure 13**) directions, then polished and etched with Keller's reagent. Recrystallization transforms the cold-worked grain structure of elongated grains into approximately equiaxed grains. However, cold rolling aligns and compresses grain boundaries in the rolling direction. This can impede recrystallization in the direction normal to the rolling direction so that grains are somewhat elongated when viewed in the longitudinal and short transverse directions as shown in **Figures 11–13**.

Figure 7 illustrates that the grain size in as-spray-rolled 2124 is uniform throughout the material regardless of G/M. Solution-heat-treated material, however, exhibits a somewhat finer grain size near the rolling surface than in the center of the strip, as shown in **Figure 8a**. This correlates with the higher concentration of constituent particles found near the surface. With increasing amounts of cold rolling following solution heat treatment, the grain size distribution becomes more uniform following recrystallization as shown in **Figure 14.** Cold reduction does not appear to appreciably change the size or distribution of secondary phase particles.

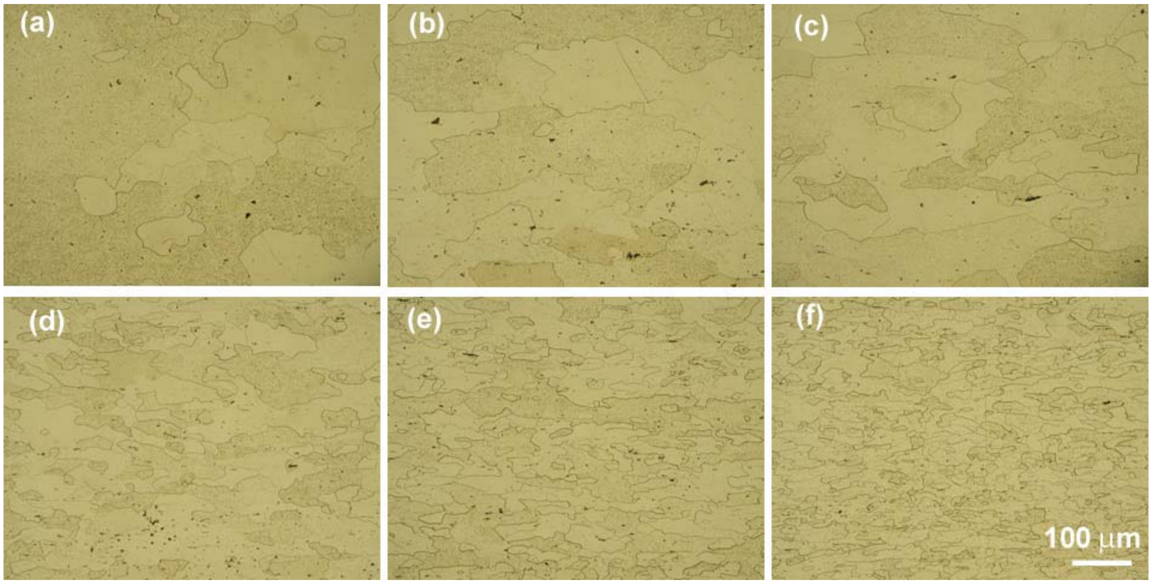

Fig. 11: Longitudinal section photomicrographs of spray-rolled 2124. Samples were solution heat treated (493°C, 2 h), cold rolled, and annealed (493°C, 10 min). Cold-roll reductions were (a) 0%, (b) 15%, (c) 30%, (d) 45%, (e) 60%, and (f) 75%.

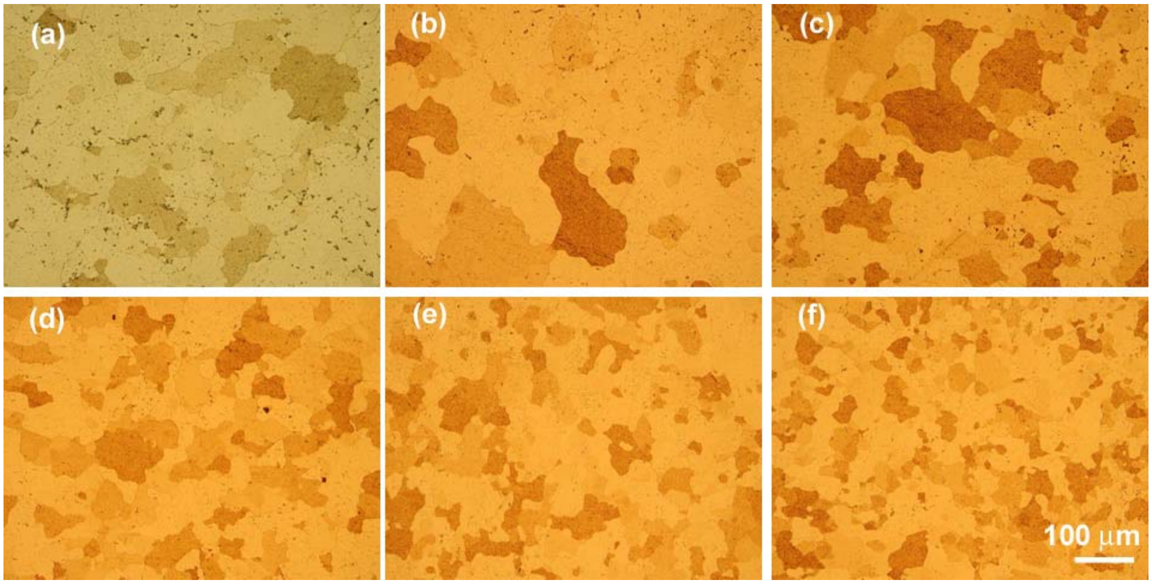

Fig. 12: Long transverse section photomicrographs of spray-rolled 2124. Samples were solution heat treated (493°C, 2 h.), cold rolled, and annealed (493°C, 10 min). Cold-roll reductions were (a) 0%, (b) 15%, (c) 30%, (d) 45%, (e) 60%, and (f) 75%.

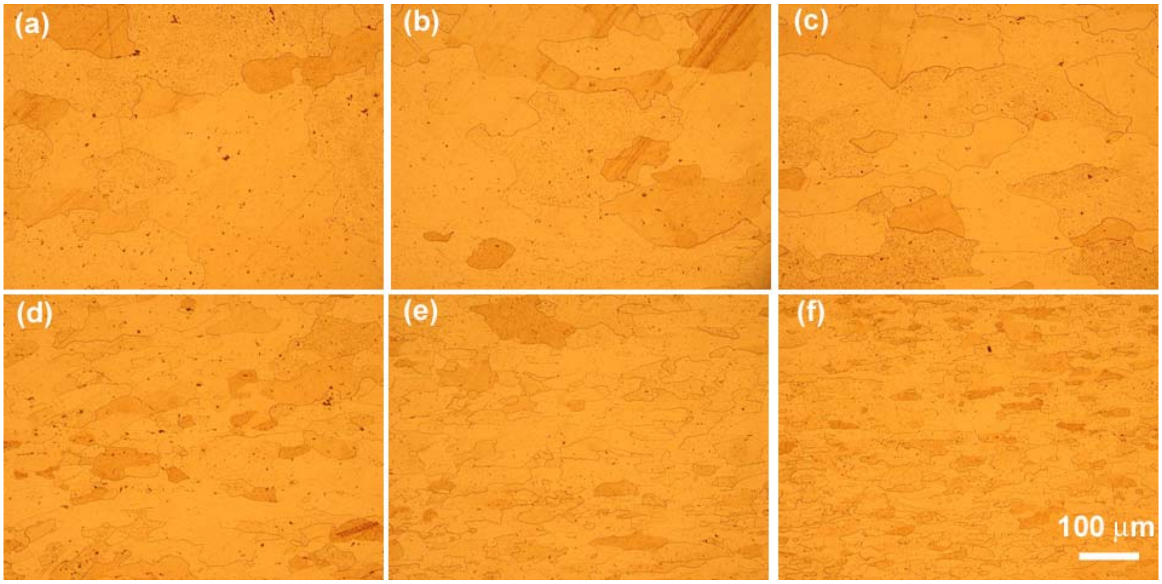

Fig. 13: Short transverse section photomicrographs of spray-rolled 2124. Samples were solution heat treated (493°C, 2 h.), cold rolled, and annealed (493°C, 10 min.). Cold-roll reductions were (a) 0%, (b) 15%, (c) 30%, (d) 45%, (e) 60%, and (f) 75%.

Fig. 14: Recrystallized grain structure near rolling surface of spray-rolled 2124. Sample was solution heat treated (493°C, 2 h.), cold rolled 75%, and annealed (493°C, 10 min.). Longitudinal section.

Conclusions

1. Spray rolling is a new strip/sheet/plate casting technology that shows promise for processing a wide variety of aluminum alloys. By combining convection cooling of atomized droplets with conduction cooling at the rolls, aluminum alloys with wide freezing ranges, such as AA2124, can be processed at high production rates.

2. The quality of strip exiting the spray-roll strip caster is sensitive to the solid fraction of the semi-solid material introduced to the rolls. Too much liquid in the semi-solid material results in poor distribution of secondary phases, which are difficult to redistribute through post-deposition TMP. With an appropriate solid fraction, strip tensile properties meet or exceed those of strip processed by the conventional ingot practices while eliminating ingot casting, homogenization, and hot-rolling unit operations.

Acknowledgments

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References

[Gra05] Ch. Gras, M. Meredith, and J. D. Hunt: *Microstructure and texture evolution after twin roll casting and subsequent cold rolling of Al-Mg-Mn aluminum alloys*, J. Mat. Process. Technol. 169 (2005) 156-163.

[McH04] Kevin M. McHugh, J. -P. Delplanque, S. B. Johnson, E. J. Lavernia, Y. Zhou and Y. Lin: *Spray rolling aluminum alloy strip*, Mater. Sci. Eng. A 383(1), 96 (2004).

[McH05] K. M. McHugh, Y. Lin, Y. Zhou, E. J. Lavernia, J. -P. Delplanque, and S. B. Johnson: *Spray Rolling Aluminum Alloy Strip For Transportation Applications*, Materials Processing and Manufacturing Division. Sixth Global Innovations Proceedings: Trends in Materials and Manufacturing Technologies for the Transportation Industry and Powder Metallurgy Research and Development in the Transportation Industry. Edited by T. R. Bieler, J. E. Carsley, H. L. Fraser, J. W. Sears, and J. E. Smugersky, TMS, San Francisco, CA, February 13-17, 2005. PP. 219-224 (2005).

[Nai06] Naiyu Sun, Burton R. Patterson, Jaakko P. Suni, Eider A. Simielli, Hasso Weiland, and Lawrence F. Allard: *Microstructural evolution in twin-roll cast AA3105 during homogenization*, Mat. Sci. Eng. A 416 (2006) 232-239.

[Yun00] M. Yun , S. Lokyer, and J. D. Hunt: *Twin roll casting of aluminum alloys*, Mat. Sci. Eng. A 280 (2000) 116-123.