Designing Aluminum Alloys for a Circular Economy

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Abstract

Aluminum is a circular metal since it can be infinitely recycled with minimal loss and degradation of its original properties. Furthermore, recycling aluminum can reduce energy consumption and CO_2 emissions by 95% compared with the primary metal extraction, leading to significant economic and environmental benefits. The use of recycled aluminum alloys in structural applications will improve the sustainability of the aluminum and manufacturing industries in a circular economy. However, the mechanical and corrosion properties of secondary (recycled) aluminum alloys are often degraded by their high "impurities" accumulated in the use phase and recycling process. This presentation presents a microalloying strategy in designing secondary alloys with higher tolerance of impurity such as iron for structural die cast and extrusion alloys.

Introduction and Motivation

Aluminum is a major lightweight material used in the transportation industries for energy efficiency.[1] However, primary production of aluminum is very energy-intensive with extensive CO_2 emission, due to the electrolytic reduction process. Re-melting aluminum only takes ~5% of the energy (and reduced emission) required to produce primary aluminum from ores.[2] The volume of aluminum scrap available for recycling is projected to double by 2050, providing a huge opportunity for making aluminum a totally circular metal, following the circular economy principles.[3] Many manufacturers, especially in the transportation industries, have committed to the use of 100% recycled aluminum in vehicle manufacturing.

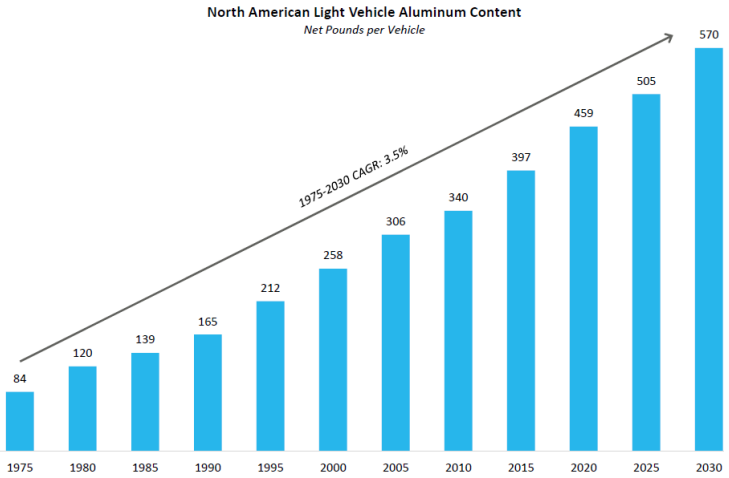


Fig. 1. Aluminum content in North American light vehicles (pounds per vehicle).[4]

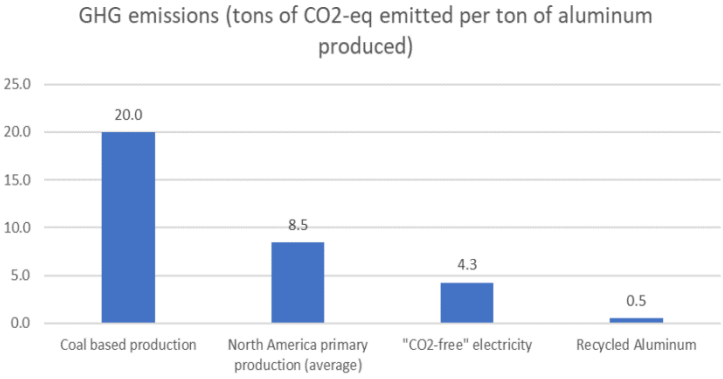


Fig. 2. GHG emissions of primary aluminum production vs. recycling. [5]

As shown in Fig. 1, the aluminum content in North American vehicles has seen significant growth in the last four decades and is expected to continue the trend in the future, especially for electric vehicles.[4] Cast and wrought aluminum alloys are increasingly used for automotive structures and closures. Typically, these alloys are *made from primary aluminum*. Secondary aluminum alloys made from end-of-life (EOL) aluminum scrap generally contain impurities (e.g., Fe, Cu and Cr), forming ***intermetallic compounds (IMCs)*** often detrimental to the alloy mechanical properties. Therefore, they are mostly used for non-structural casting

applications (such as internal combusion engine and transmission components), not for structural applications in automotive body and chassis systems.

The recycling of aluminum scrap not only helps reduce landfill waste but also plays a crucial role in significantly lowering greenhouse gas (GHG) emissions (Fig. 2[5]) of aluminum in the circulation process. To achieve this vision, new secondary alloys with improved mechanical properties (at least matching the primary alloys) would provide enormous economic and environmental benefits. This paper presents a microalloying strategy in designing secondary alloys with higher tolerance of impurity such as iron for structural die casting and extrusion applications.

Review of Related Work

The current approach in structural aluminum castings and wrought products is restricting impurity content such Fe to within about 0.2%[1], which limits the use of EOL automotive scrap known as "Twitch." Fig. 3 illustrates the process flow for recovering aluminum scrap (Twitch) at the EOL stage of automobiles.[6] The process starts with a scrapped vehicle at a dismantling facility, where reusable components and hazardous materials are removed. The vehicle is then flattened and shredded. Other post-consumer scrap streams are typically mixed with the EOL shred. The shredded material undergoes sorting through magnetic, air, and Eddy-current separators to isolate non-ferrous auto shred, known as Zorba. Most Zorba is exported, while the portion that remains domestic is processed further using density separation to produce Zebra and Twitch fractions. Zebra refers to heavier non-ferrous scrap, mostly copper and zinc, while Twitch is a light non-ferrous scrap primarily consisting of aluminum with small allowances of Mg, Zn, and Zn alloys. Fig. 4 presents aggregated data on the average chemical composition of melted Twitch, collected from both East and West Coast streams over three quarters in 2023.[6] It shows an average Fe content of 0.57±0.04 wt.% and Mn of 0.23±0.02 wt.% among other elements.

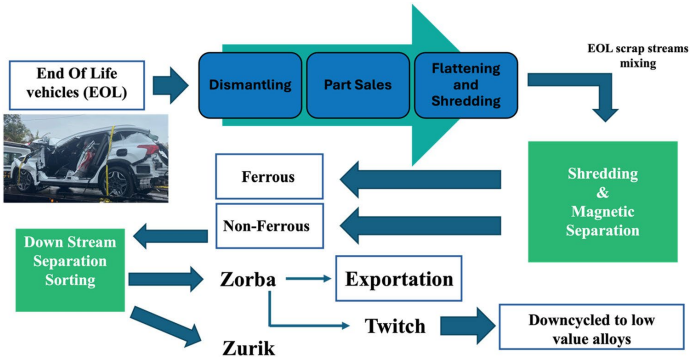


Fig. 3. Process flow for the recovery of aluminum scrap (Twitch) from EOL automobiles. [6]

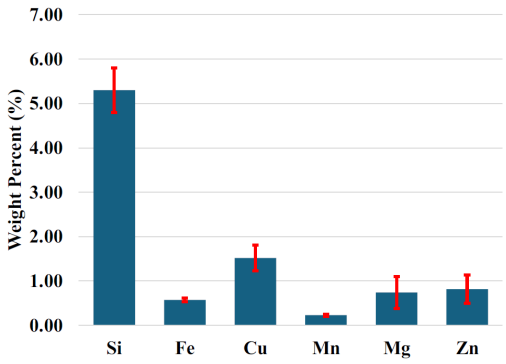


Fig. 4. Average elemental composition of Twitch; data from 3 quarters in 2023, east and west coast streams. [6]

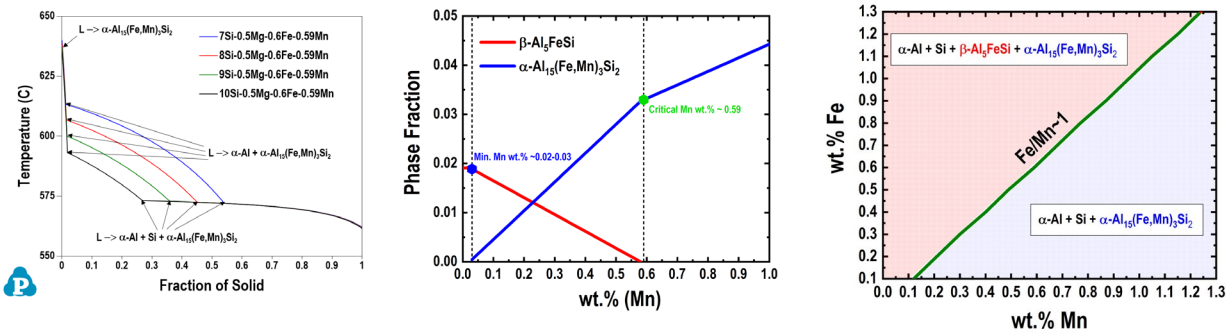
Technology Approach

As discussed above, the major challenge in designing recycled aluminum alloys from Twitch scrap is the control of IMCs in the alloy microstructure. The fundamental approach discussed in this paper is to control/optimize IMC formation via microalloying to suppress/minimize undesirable IMC phase formation in final alloy microstructure.

The CALPHAD (CALculation of PHAse Diagrams) approach [7] has been developed to predict phase formations in an alloy system with thermodynamic parameters predefined for all possible phases in thermodynamic databases. It can be used to model the microalloying effect on the formation of IMCs in multicomponent secondary alloys based on Twitch materials with high Fe contents. Fig. 5 is an example of how the CALPHAD approach is used to predict IMC formation in secondary aluminum alloy. [8] Fig. 5a shows a high throughput calculation (HTC) of solidification paths of Al-7Si-0.5Mg-0.6Fe alloy with various Mn additions. Fig. 5b shows the total phase fractions of β-Al_5FeSi and α-$Al_{15}(Fe,Mn)_3Si_2$ phases vs. Mn content for Al-8Si-0.35Mg-0.6Fe. A formation map of Fe-containing intermetallic phases was plotted for Mn vs. Fe contents, which defines a critical Fe/Mn ratio of 1 for α-$Al_{15}(Fe,Mn)_3Si_2$

[1] All compositions are in wt.% except otherwise stated.

vs. β-Al_5FeSi (Fig. 5c). It is well known that β-Al_5FeSi is significantly more detrimental than α-$Al_{15}(Fe,Mn)_3Si_2$ to mechanical and corrosion properties of aluminum alloys.



a. Calculated solidification paths of Al-7Si-0.5Mg-0.6Fe alloy with various Mn additions.

b. Phase fraction of Fe-containing intermetallic phases with Mn microalloying.

c. Effect of Fe and Mn contents on the formation of intermetallic phases.

Fig. 5. CALPHAD modeling of Fe-containing intermetallic phase formation in aluminum alloys. [8]

Discussion

Microalloying

The influence of Sr and cooling rate on the formation of Fe-containing intermetallic phases during solidification was examined in Al-9Si-0.6Fe-0.35Mg-(0.1-0.3)Mn alloys, with the findings summarized in Fig. 6.[8, 9] The additions of 60 and 200 ppm Sr refine Si needles, transforming their morphology from coarse, flake-like structures to a lamellar form, while also influencing the formation of β−AlFeSi phases. In Sr-modified alloys, the needle-shaped β−AlFeSi phase is characterized as the δ-Al_3FeSi_2 with an Fe-to-Si ratio of 1:2, and this morphology remains largely unchanged under low cooling rates (1.5 °C/s). However, at the fastest solidification cooling rate of 60 °C/s, the addition of 60 ppm Sr significantly reduces the length of the needle-shaped δ-$AlFeSi_2$ phase, confining it within eutectic regions with an average length of 3–5 µm. Increasing Sr to 200 ppm leads to longer δ-Al_3FeSi_2 needles, ranging from 10 µm to over 50 µm.

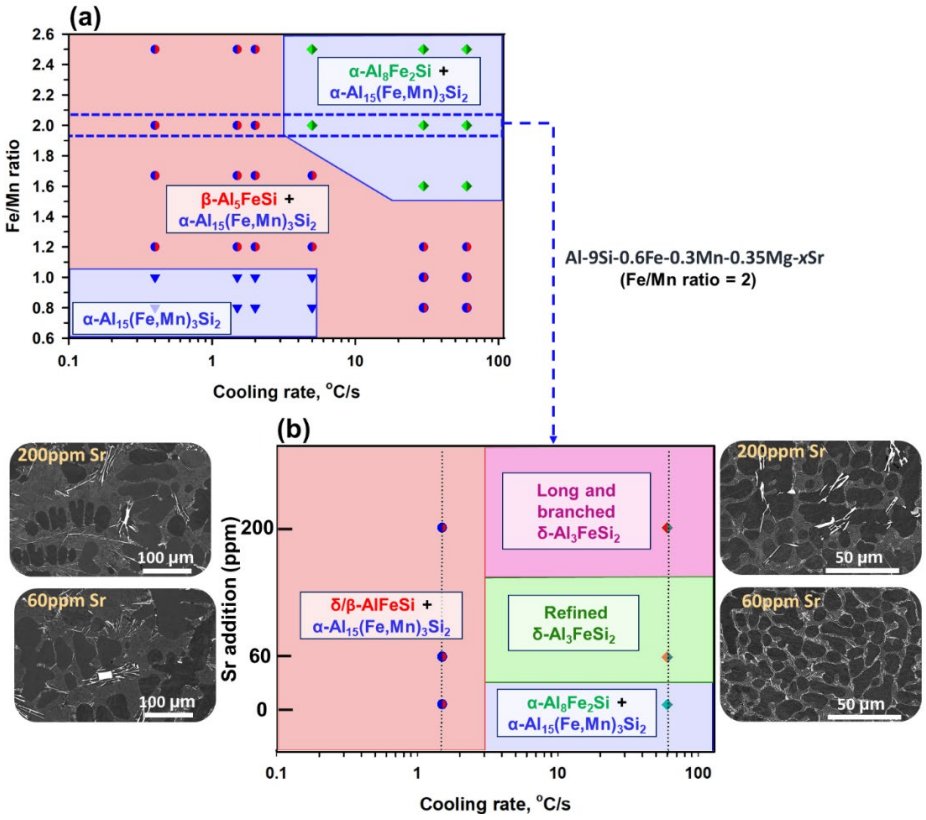


Fig. 6. Phase formation map of Fe-based intermetallics reported previously for (a) Al-9Si-0.35 Mg-xFe-xMn alloys (adapted from [8]*) and (b) for the present work updated with Sr addition at Fe-to-Mn ratio of 2 in the same casting conditions.* [9]

Mechanisms underlying the impact of Sr on δ-phase formation were analyzed using CALPHAD simulations and experimental observations. At 60 ppm Sr, the refinement of δ-Al_3FeSi_2 phase is attributed to Sr poisoning nucleation

sites and reducing the phase's formation temperature. Conversely, at 200 ppm Sr, the increased fraction of Al_2Si_2Sr phase promotes early nucleation of δ-Al_3FeSi_2 phases, allowing for unrestricted growth and resulting in long, branched δ-phase needles. For scrap aluminum such as Twitch containing up to 0.6% Fe, a 60 ppm Sr addition and a high cooling rate (such as die casting) provides optimal refinement of needle-shaped eutectic Si and δ-Al_3FeSi_2 phases.

It is also reported [10] that combined microalloying of Mn and Cr to Al-Si-based alloys can be more effective in suppressing the harmful platelet-type $Al_xFe_ySi_z$ intermetallic phases into the less detrimental script or polygonal α-$Al_x(Fe,Mn,Cr)_ySi_z$ phase. The morphology, composition, and behavior of α-$Al_x(Fe,Mn,Cr)_ySi_z$ are influenced by solidification conditions. Figure 7 illustrates the morphological evolution and underlying mechanisms of α-$Al_x(Fe,Mn,Cr)_ySi_z$ in an Al-Si-Fe-0.2Cr alloy with Cr additions. Thermodynamic simulations using the CALPHAD method were conducted to compare phase compositions and formation temperatures of the α-$Al_x(Fe,Mn,Cr)_ySi_z$ phase with experimental results. Findings revealed that adding 0.25Mn and 0.2Cr to an Al-9Si-1Fe alloy results in compact polygonal phases at high cooling rates (i.e., die casting), whereas gravity castings predominantly exhibit mixed structures of platelets and script-type morphologies. Tensile results showed that the mixed structure of platelets and script phases achieved a higher elongation of 6.6%, while the polygonal phases resulted in a reduced elongation of 4.2%. This study underscores the critical role of solidification conditions in shaping the formation of Fe-containing intermetallic phases and their corresponding mechanical properties. It also draws comparisons with relevant literature to highlight the implications for high-pressure die-casting process.

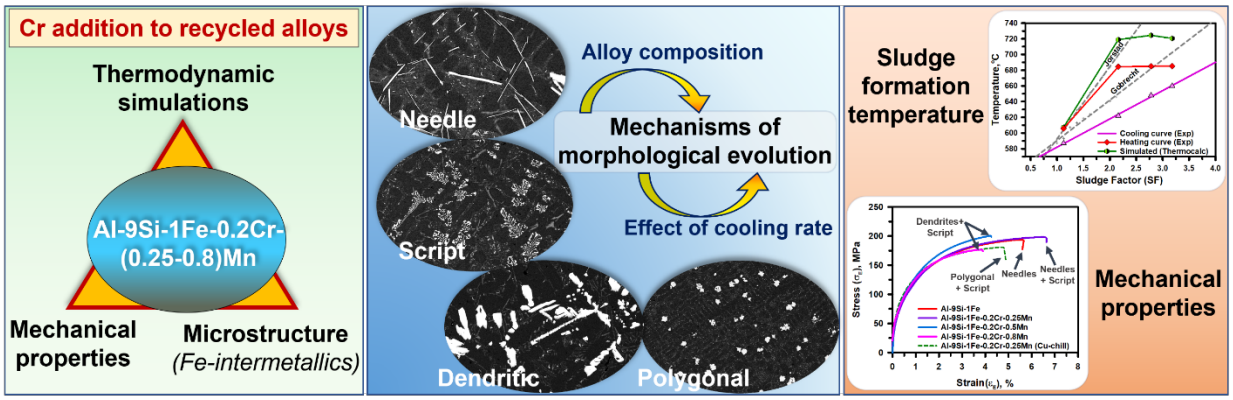


Fig. 7. Controlling the Fe-intermetallic phases and mechanical properties of secondary Al-9Si-1Fe alloy with Cr and Mn additions. [10]

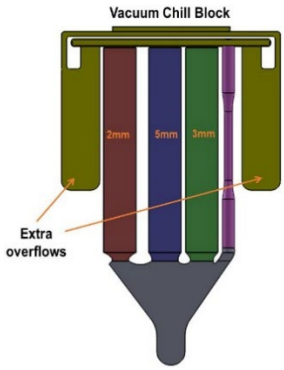
Alloy Design for Die Casting [11]

Based on the above thermodynamic simulation results, two structural alloys, RS-1 (containing 0.5%Fe) and RS-2 (containing 0.6%Fe), have been designed with microalloying elements Mn, Cr and Sr (see Table 1 for compositions). Three non-structural alloys, NS-1, NS-2, and NS-3, have also been designed with a base composition Al-7Si-0.4Mg-1.8Cu-0.9Zn-1Fe and some microalloying elements (see Table 2 for compositions). It should be noted that the base composition for non-structural alloys have higher Mg but lower Cu content than A380 to reduce cost (no/minimal Cu addition) and environmental impact (eliminating chlorination step of Mg removal) during recycling.

The above experimental alloys (made from 100% Twitch scrap aluminum by Audubon Metals), along with EZCast C611 (baseline for structural alloys, supplied by Alcoa) and A380 (baseline for non-structural alloy, supplied by Audubon Metals), were die cast for property evaluation. A 280-ton LK die casting machine (Fig. 4a), equipped with a Fondarex vacuum unit at The Ohio State University, was used for the die casting trials. An experimental die design was employed (Fig. 4b), featuring test plates with thicknesses of 2, 3, and 5 mm, along with an ASTM standard round bar (6 mm diameter in the gauge section). The specimens were produced using a vacuum-assisted die casting process, with the die cavity vacuum level set at 85 mbar, to minimize gas porosity from entrapped air. The die was preheated to 200 °C, and the shot sleeve was maintained at 270 °C. Molten alloys, heated to 680–730 °C depending on the alloy, were transferred to the shot sleeve using a robotic ladling arm and a boron-nitride-coated steel ladle. The low die temperature (200 °C) was used to enhance cooling rate for refined microstructure. For each alloy, about 400 lbs. of melt was used to make at least 100 shots (see Fig. 4c for an example casting) for testing and evaluation.

Table 1 shows the major compositions and tensile properties of recycled structural die cast alloys. For structural alloys, both RS-1 (0.5Fe) and RS-2 (0.6Fe) alloys have shown similar ductility (around 10% elongation), slightly higher yield and ultimate tensile strengths than those the baseline EZCast 611 alloy, but at significantly higher Fe content (0.5-0.6%) accommodating 100% scrap aluminum in RS-1 and RS-2 alloys. Table 2 shows the major compositions and tensile properties of recycled non-structural die cast alloys (in comparison with baseline A380 alloy). For non-structural alloys, NS-1, NS-2 and NS-3 alloys show slight increases in elongation and similar strength levels, compared with the baseline A380 alloy. It should be noted that NS-1, NS-2 and NS-3 alloys have higher Mg but lower Cu content compared to A380. Those recycled alloys would eliminate the de-Mg operation (using chlorine gas) and save Cu additions during the recycling process, thus, have lower cost and as well as being more environmentally friendly.

(a) 280 ton die casting machine



(b) Specimen die design

(c) Example casting

Fig. 8. Die casting trials of recycled aluminum alloys and baseline alloys at Ohio State University.

Table 1. Major compositions and mechanical properties of recycled structural die cast alloys (in comparison with baseline EZCast C611 alloy)

Alloy	Temper	% Elongation	YS (MPa)	UTS (MPa)	Sample
RS-1 (Al-9Si-0.3Mg-0.3Cu-0.5Fe + microalloying)	F	7.42	123	273	Round (6 mm)
	F	7.45	96	292	Plate (3 mm)
	T7*	10.1	103	214	Plate (3 mm)
RS-2 (Al-7Si-0.3Mg-0.5Cu-0.6Fe + microalloying)	F	7.6	125	275	Plate (3 mm)
	T7*	9.8	117	219	Plate (3 mm)
EZCast C611 (Al-7Si-0.2Mg-0.1Fe)	*F*	*7.9*	*105*	*228*	*Round (6 mm)*
	F	*8.9*	*99*	*235*	*Plate (3 mm)*

*T7 Heat treatment procedure: 490°C for 48 min followed by forced air quench to 80-100°C, 230°C for 96 min followed by natural air cooling.

Iron Tolerance Limit in Extrusion Alloys

Automotive-grade aluminum extrusions are currently produced using primary aluminum, which has a higher carbon footprint than recycled alloys. A recent paper [12] outlines a pathway to significantly reduce the carbon footprint of automotive-grade aluminum extrusions by utilizing EOL scrap (Fig. 9). Thermodynamic simulations (Fig. 10) determined an iron tolerance limit of 0.25% for AA6082 extrusion. This allows for a scrap usage of more than 70% during billet casting, which was successfully validated in a production trial. The resulting low-carbon AA6082 alloy extrusions exhibited a yield strength of 316.3 ± 3.5 MPa, an ultimate tensile strength of 343.9 ± 1.9 MPa, and a tensile elongation of 13.4 ± 1.2%, all comparable to the performance of commercial AA6082 extrusions made from 100% primary aluminum. Additionally, component-level 3-point bending tests of front bumper beams made from both low-carbon and commercial AA6082 alloys demonstrated equivalent performance. These findings suggest that upcycling EOL aluminum scrap to produce aluminum extrusions is a viable strategy for decarbonizing the vehicle manufacturing process.

Table 2. Major compositions and mechanical properties of recycled non-structural die cast alloys (in comparison with baseline A380 alloy)

Alloy	% Elongation	YS (MPa)	UTS (MPa)	Sample
NS-1 (Al-7Si-0.4Mg-1.8Cu-0.9Zn-1Fe + microalloying 1)	2.5	161	273	Plate (3 mm)
	3.4	160	288	Plate (5 mm)
	2.2	148	259	Round (6 mm)
NS-2 (Al-7Si-0.4Mg-1.8Cu-0.9Zn-1Fe + microalloying 2)	4	130	266	Plate (3 mm)
	4.4	128	266	Plate (5 mm)
NS-3 (Al-7Si-0.4Mg-1.8Cu-0.9Zn-1Fe + microalloying 3)	3.0	161	277	Plate (3 mm)
	4.0	155	294	Plate (5 mm)
	3.7	142	272	Round (6 mm)
A380 (Al-9Si-0.1Mg-3.4Cu-1Zn-0.9Fe-0.3Mn-0.04Cr)	*2.1*	*151*	*236*	*Plate (3 mm)*
	2.5	*144*	*228*	*Plate (5 mm)*
	2.7	*136*	*281*	*Round (6 mm)*

It should be pointed out that the properties (Tables 1 and 2) obtained from die cast test specimens still need to be further validated in component-level testing, which will be conducted in a future project led by industrial partners of this project.

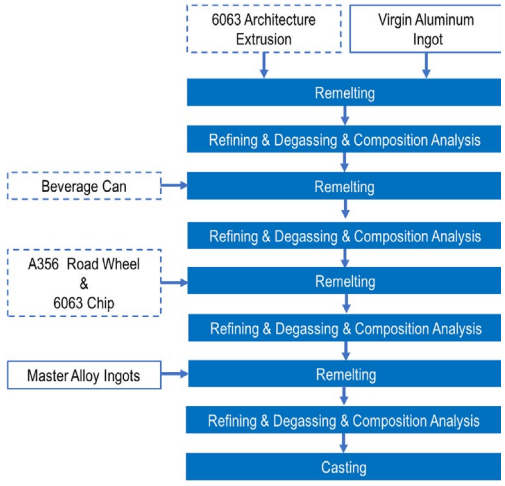


Fig. 9. Material flow chart including recycled content in aluminum extrusion alloys. [12]

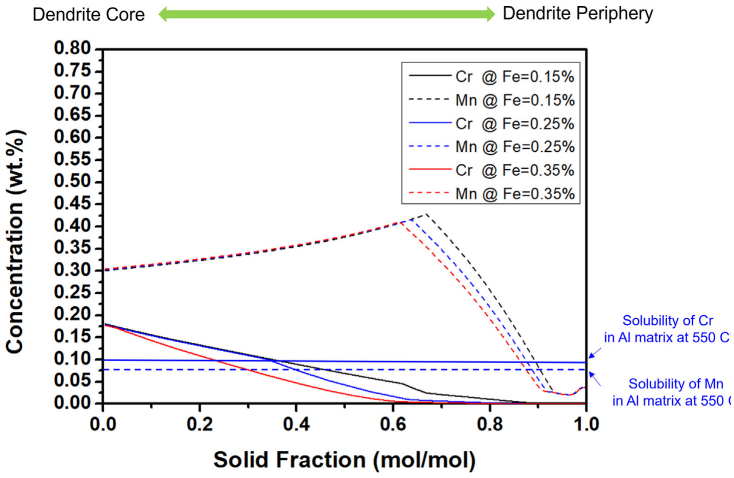


Fig. 10. Cr and Mn concentrations in the crystallized Al solid as a function of the solid fraction (Scheil solidification simulation). [12]

Conclusions and Recommendations

Increased use of recycled aluminum (both cast and wrought products) in the transportation industry will not only save energy and reduce greenhouse gas emissions due to vehicle lightweighting but improve sustainability via reduced primary aluminum production, contributing to a net-zero and circular economy. Microalloying is an effective approach in controlling/optimizing intermetallic phases to suppress/minimize undesirable IMC phase formation in final alloy microstructure and increase the recycling content in structural casting and extrusion alloys.

Acknowledgements

The authors acknowledge the contributions of their colleagues and students at The Ohio State University, University of California Irvine, and many industry partners including Alcoa, Audubon Metals, CompuTherm, Ford Motor

Company, General Motors, North American Die Casting Association, The Aluminum Association, and American Foundry Association. “Part of this research is supported by the U.S. Department of Energy’s Office of Energy Efficiency and Renewable Energy (EERE) under the Advanced Manufacturing Office Award Number DE-EE0007897, awarded to The REMADE Institute, a division of Sustainable Manufacturing Innovation Alliance Corp. This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.”

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