SOLID-STATE UPCYCLING OF ALUMINUM ALLOYS

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Abstract

Aluminum alloys are crucial materials in automotive and aerospace manufacturing sectors due to their high specific strength, corrosion resistance, and lightweight properties, which provide better fuel efficiency and mechanical performance. Extraction of Al from bauxite ore is an energy intensive process. Conventional aluminum recycling involves several steps: scrap collection, shredding, cleaning, melting, casting, and rolling. While this process helps reduce the need for raw materials and energy compared to ore extraction for producing primary aluminum, it still involves substantial energy consumption and has major environmental drawbacks. Particularly, melting and casting require significant energy inputs and produce significant greenhouse gas emissions. The International Aluminum Institute (IAI) estimates that the primary Al production is expected to reach a volume of 82 million tons by 2050 whereas Al recycling is expected to grow at a rather higher rate to hit 70 million tons, which is ~30% higher than the present primary Al to recycled Al ratio. Additionally, IAI also estimates the primary Al production to decline and reach today's rate of production whereas Al-recycling is expected to increase to 120 million tons by 2070. Such a future trend in Al-recycling tonnage demands exploration of novel approaches that align with the principles of circular economy and sustainability. One such alternative is to recycle Al without melting. Friction stir based processes are ideal for this. An overview of our upcycling effort using SolidStir® extrusion and SolidStir® additive manufacturing will be presented. Our analysis has shown that the energy requirement for Al-recycling using the SolidStir® technology can be substantially reduced to 0.78 kWh/kg, which is mere 1.7% of energy required for primary aluminum extraction and only 28% of the energy required for conventional Al recycling. The results demonstrate substantial energy saving compared to the conventional recycling route (72% less), and mechanical properties exceeding the base alloy properties.

1. Introduction

Aluminum alloys have emerged as crucial materials in automotive and aerospace manufacturing sectors due to their high specific strength, corrosion resistance, and lightweight properties, which provide better fuel efficiency and mechanical performance [1]. Extraction of Al from bauxite ore being an energy intensive process, recycling of Al alloys which requires 95% less energy is sought to reduce ecological footprints [2]. In the US, ***~3.4 billion lbs of aluminum alloys are estimated to get recycled each year***, with the transportation industry leading the way, accounting for about 33% of this total. Approximately one third of present aluminum alloy production is from recycled scrap which is expected to increase by 50% in 2050 [3].

Conventional aluminum recycling involves several steps: scrap collection, shredding, cleaning, melting, casting, and rolling (Fig. 1) [4,5]. While this process helps reduce the need for raw materials and energy compared to ore extraction for producing primary aluminum, it still involves substantial energy consumption and has major environmental drawbacks. Particularly melting and casting, requires significant energy inputs and can produce greenhouse gas emissions [3]. Improving and optimizing recycling processes is crucial for reducing their overall environmental impact. This necessitates the exploration of novel approaches that align with the principles of circular economy and sustainability. ***One such alternative is to recycle Al without melting***. Additional opportunity comes from being able to convert the aluminum alloy scrap into aluminum-matrix composite. Some scrap is labeled as "dirty" due to the presence of Fe-rich contaminants, and the ***paradigm shift will be to take a dirty-scrap and make it dirtier by adding***

second phase particles which will result in ***enhanced performance through composite strengthening***.

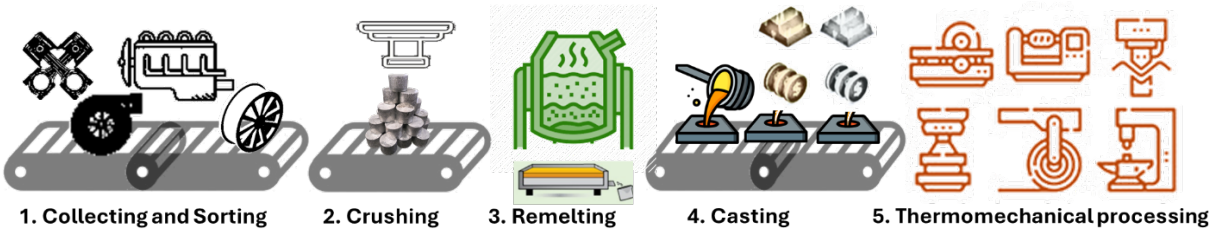

Figure 1: Steps involved in conventional Al recycling before any subtractive manufacturing.

2. Aluminum recycling: state-of-the-art

Aluminum production from ore is highly energy intensive which accounts for ~3% of greenhouse gas (GHG) emissions, equivalent to 12-16.5 tons of GHG per 1 ton of Al produced. Surprisingly, being an infinitely recyclable material, 75% of Al produced in history is still in use. The International Aluminum Institute (IAI) estimates that the primary aluminum production is expected to reach volume of 82 million tons by 2050 whereas Al recycling is expected to show a rather higher rate to hit 70 million tons, which is ~30% higher than the present primary Al to recycled Al ratio. Additionally, IAI also estimates the primary Al production to descent and reach today's rate of production whereas Al-recycling is expected to mount to 120 million tons by 2070 (Fig. 2). Such a future trend in Al-recycling tonnage demands exploration of novel, efficient and economical methods with high productivity for Al recycling. The current Al recycling involves remelting and casting of Al scrap which utilizes only 5% of the energy requirements for primary aluminum extraction [3]. However, the energy requirement is still high, and the process expels ~0.6 kg of CO_2/kg [3]. These cast recycled materials need to undergo additional metal forming processes to eliminate defects, improve mechanical properties, and provide final shapes [6]. Traditional recycling methods, notably those reliant on melting, present challenges in terms of energy consumption and potential compromises to material properties. Conventional metal recycling also consists of steps like collection, sorting, processing, melting, purification, solidification, and forming, introducing a myriad of complexity.

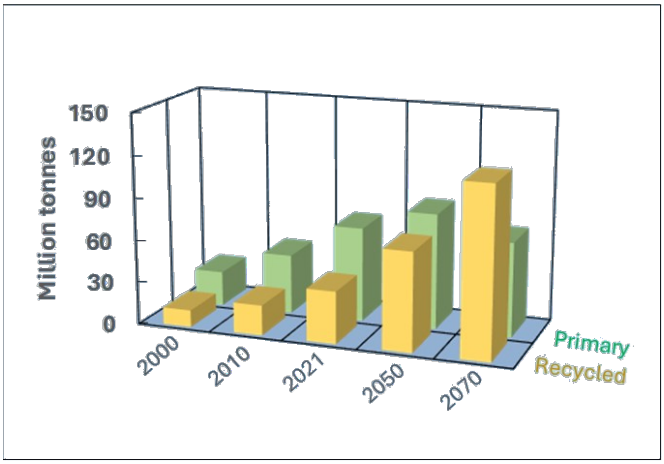

Figure 2: Statistical estimate of primary and recycled aluminum.

Conventional solid-state recycling involves Al-chip recovery by means of plastic deformation such as hot extrusion, forging, rolling, channel pressing, and friction extrusion [3,8–10]. Recently, advanced solid state additive manufacturing (AM) technique such as additive friction stir deposition (AFSD) has been utilized for aluminum and titanium recycling [4,11,12]. In such cases, the final build exhibited a wrought-like microstructure, demonstrating strength and ductility comparable to that of the base material. Notably, the shear-assisted processing and extrusion technique (ShAPE®) developed by Pacific Northwest National Laboratory has been successfully demonstrated for Al scrap recycling [13–17]. The existing solid-state recycling routes are currently end-product specific whereas ShAPE® has successfully demonstrated end-product customization. However, such processes are still limited to extrusions. ***An integrated process which enables part fabrication from recycled scrap, local repair on existing parts, and AM will***

expand end-product flexibility and customization. Such a process will have revolutionary impact on existing Al-recycling state-of-the-art, a perfect fit for SolidStir®.

3. SolidStir® technology: a candidate for sustainable manufacturing

The revolutionary invention of friction stir welding (FSW) in 1991 at The Welding Institute, UK, paved the way for various technological innovations over the last three decades [7]. Recently, friction stir principles have been utilized for advanced manufacturing processes such as solid-state extrusion and additive manufacturing [8–10]. SolidStir® extrusion (Fig. 3) is an innovative, disruptive thermo-mechanical process, pioneered by Enabled Engineering, which allows fabrication of a variety of extrudate shapes dictated by the chamber throat cross-section design. In its conventional form, SolidStir® (patented by Enabled Engineering [11]) utilizes the contact friction between a non-consumable rotating tool to plasticize the material fed into a confined die chamber.

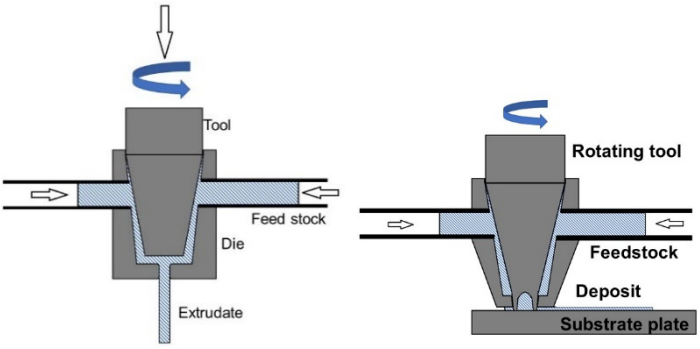

Figure 3: Schematic of extrusion and additive manufacturing modes of SolidStir®.

The plasticized material is pushed downward through the die throat customized to achieve the desired extrudate shape such as rods, wires, and tubes (Figs. 4(a-c)), with the assistance of spiral features on the tool. Control over material flow and degree of deformation is realized by modifying the tool features and process parameters. The extrusion rate for a specific cross-sectional geometry can be controlled through the feedstock feed rate and process parameters. The mechanism that feeds the material through the feed port can be varied depending on the form of feedstock material. The unique design of the technology allows flexibility in feedstock form such as machined chips and scraps to be supplied directly into the processing chamber. Also, multiple feed ports can be provided through which feedstocks can be supplied alternatively to achieve continuous extrusion [12]. Fig. 3(c) shows 75-feet of 3 mm wire extruded using the technology using multiple feedstocks of 50 mm length. In addition to being energy-efficient (requiring less energy than traditional re-melting of waste metals), eco-friendly, and a carbon-inhibitor (reduction in carbon footprint by reducing the energy requirements), SolidStir® technology can produce materials (ingots) and components (fabricated parts/builds) of similar or even better strength and properties than the original material, hence the term ***upcycling***. SolidStir® fabricated parts are near-net shape, size, and configuration which require limited machining to produce the newly designed or existing components, resulting in reduced machining cost. Apart from economic benefits and process novelty, the proposed technology provides a wrought-like microstructure to the final product with very fine recrystallized grains, reaping the benefits of the friction-stir based thermomechanical process.

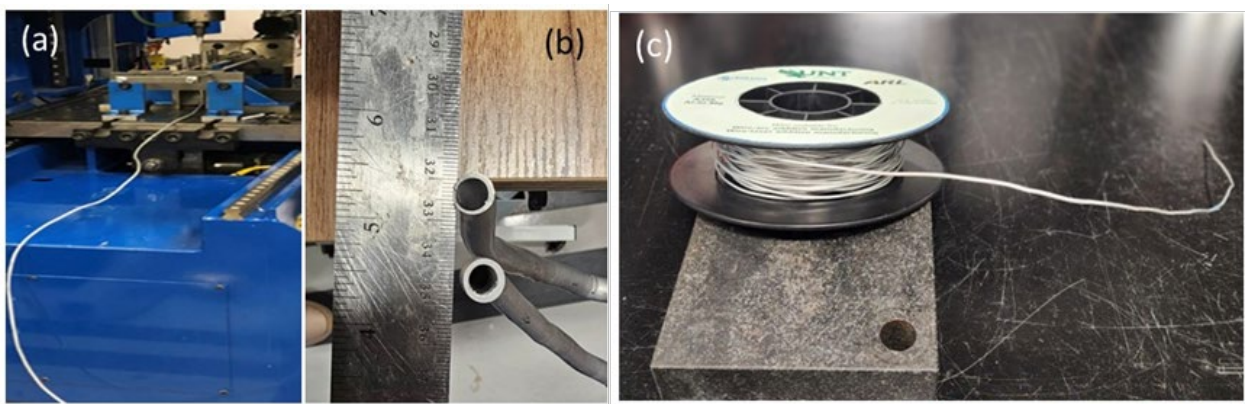

Figure 4: (a-c) Images of SolidStir® extruded circular rod (6 mm diameter), tube (10 mm outer diameter) and 75-feet wire (1.67 mm diameter).

US Environmental Protection Agency defines sustainable manufacturing as “the creation of manufactured products through economically-sound processes that minimize negative environmental impacts while conserving energy and natural resources”. Achieving sustainability for metal manufacturing requires a comprehensive approach that considers environmental, economic, and social impacts [13]. The sustainability of a metal should not only consider aspects of primary production but also its end-of-life recyclability [14]. From a product lifecycle management viewpoint, innovating appropriate recycling technologies boosts the reprocessing capabilities in designing circular economies for materials by adopting “cradle-to-gate” instead of “cradle-to-grave” strategy [15]. As the demand for metals and the associated energy requirements grow, the development of energy-efficient recycling and upcycling technologies becomes increasingly important to ensure the sustainability of critical resources [16].

At present, transportation industry contributes to maximum Al recycling in both tonnage and recycling efficiency rate (RER). The global Al recycling tonnage by transportation industry accounts for 9 million tons with 86% global RER [17]. Reports also show that the RER in certain automobile companies even reaches 95% [17]. The metal waste associated with automobile industry comes from multiple sources (Fig. 5). There are two important aspects of the scrap, (a) composition, and (b) form (size and shape of the scrap). A major challenge arises from mixed scrap where different aluminum alloys are inadvertently mixed. For example, if a machining center is collecting scrap during machining of a variety of components without any regards to separation of various grades, then the resulting scrap produces challenges during recycling. Therefore, there are two needs. First, to establish protocols to ensure proper scrap generation by being mindful of the circular manufacturing. Second, research towards redefining of recycling technologies play a vital role in continual evolution of the manufacturing sector, ensuring sustainable growth with minimal environmental impact [18]. Hence, development of new technologies should ensure environmental sustainability, economic benefits, and material performance. The innovation of SolidStir® promotes the "***Reduce, Reuse, Recycle, Redesign, Recover, and Remanufacture***" framework to promote sustainable manufacturing with minimal environmental impact. The process needs to be robust to create proper metallurgical microstructure that shields any harmful effect of unintended contaminants.

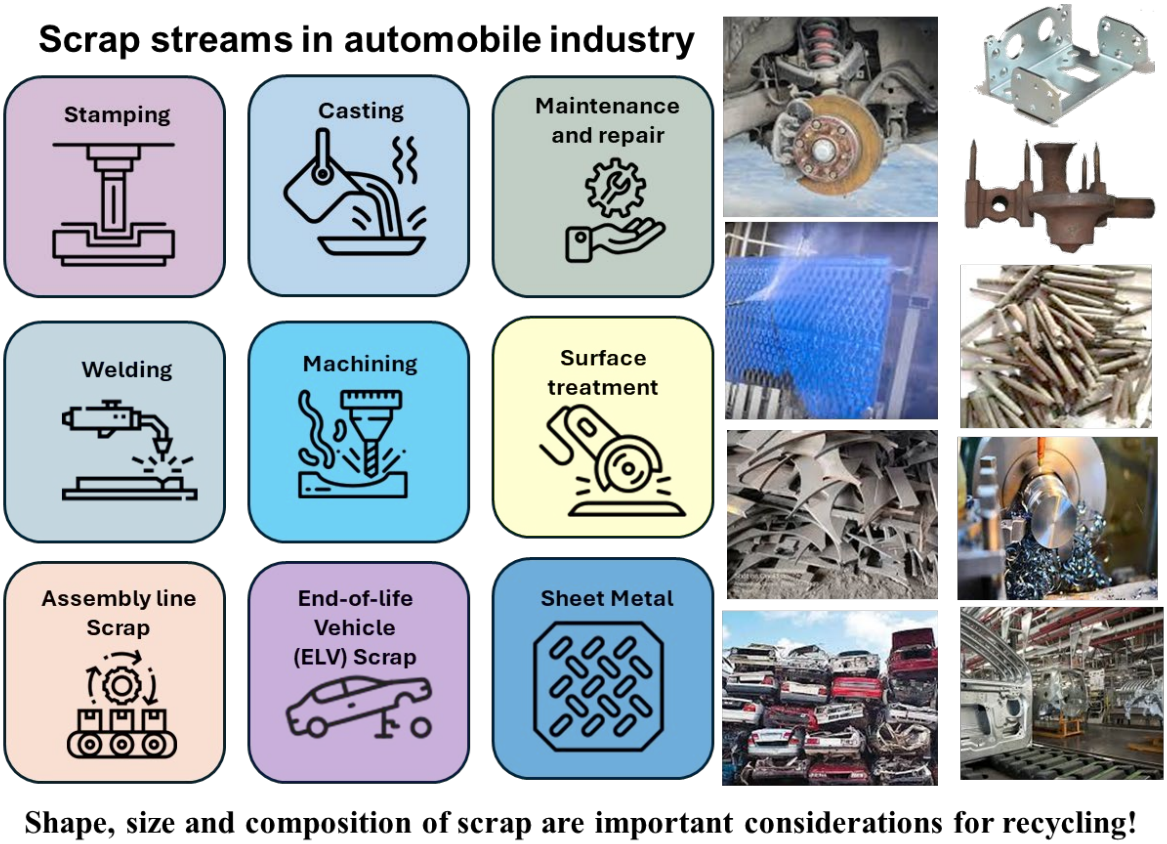

Figure 5: Sources of metal scrap in an automobile production and assembly.

4. Solid-state recycling of aluminum alloys

Solid-state recycling of aluminum alloys presents a sustainable and energy-efficient alternative to traditional melting-based recycling methods. By ***eliminating the need for high-temperature melt processing, this approach significantly reduces energy consumption and minimizes greenhouse gas emissions, contributing to a lower carbon footprint***. Additionally, solid-state recycling preserves the original material properties by preventing oxidation and the loss of alloying elements, which are common in melting processes. The reduced metal loss and contamination risks further enhance the quality and recovery rates of recycled aluminum. Moreover, the process generates fewer harmful by-products, such as dross and toxic gases, making it environmentally friendly. With shorter processing times, cost-effectiveness, and suitability for a wide range of aluminum scraps, solid-state recycling offers a versatile and scalable solution, particularly for industries prioritizing resource efficiency and waste minimization.

Figure 6 illustrates the application of the newly developed solid-state recycling technology, SolidStir® extrusion, to recycle machined aluminum (AA 356 and AA 5083) chips. To prevent metallurgical issues associated with contamination, dry aluminum chips were machined from a cast block without usage of lubricant. These chips were then compacted into cylindrical pellets measuring 10 mm in diameter and approximately 1 inch in length using a hydraulic press. Multiple compacted chip pellets were subsequently fed into the SolidStir® system, producing a continuous 6 mm diameter extruded rod, as shown in Figure 6. The shape and size of the extrudate can be changed through the design of the die opening. In the proof-of-concept stage, a simple circular shape of the extrusion was selected.

The processed rod underwent microstructural characterization and mechanical testing. Microstructural analysis was performed using a scanning electron microscope (SEM) (ThermoFisher Scientific Apreo-2) equipped with an electron backscatter diffraction (EBSD) detector (Velocity-Pro). EBSD data acquisition and analysis were carried out using EDAX APEX and TSL OIM Analysis™ 8 software, respectively. Tensile testing was conducted at room temperature on rectangular dog-bone-shaped mini-tensile specimens (gauge length: 5 mm, width: 1.25 mm, thickness: 1 mm) using a custom-built mini-tensile tester, with an initial strain rate of 10^{-3} s^{-1}. Fractography was performed by capturing micrographs of the fractured surfaces.

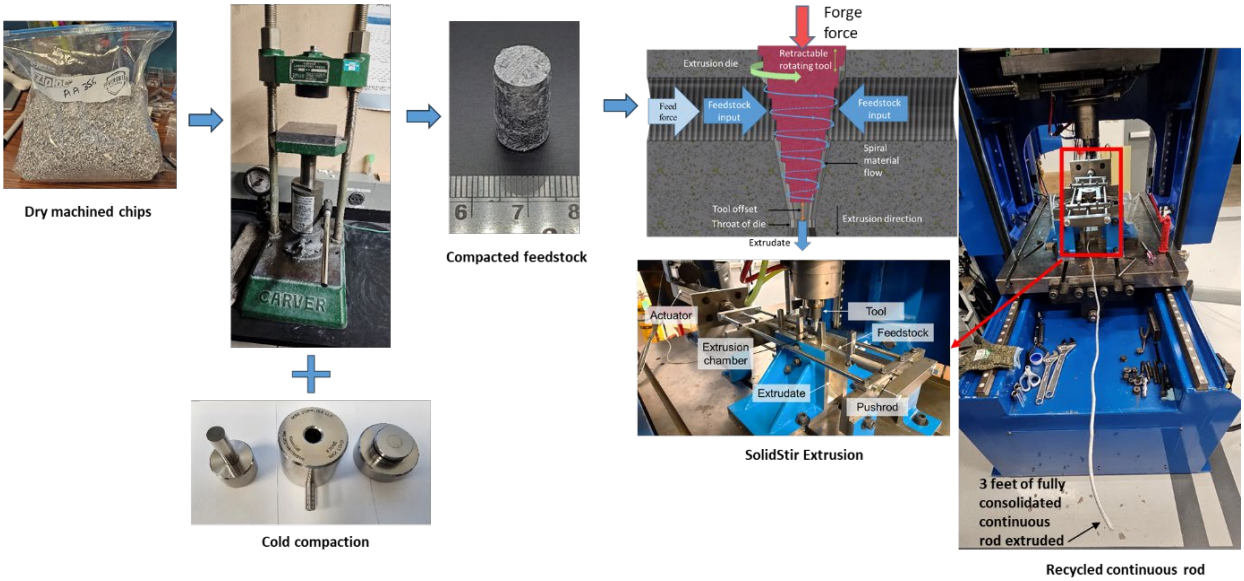

Figure 6: Dry machined chips were compacted to prepare feedstock for recycling using SolidStir® Extrusion to make a continuous extrudate.

4.1. Microstructural Analysis

Figure 7 exhibits the microstructure of the AA356 base material. Note that AA356 alloy is a casting alloy and the microstructure consists of silicon and iron rich particles which are large in size (30-100 µm). During SolidStir® recycling these particles were sheared (to a size of <5 µm) and distributed uniformly in the aluminum matrix. The recycled material has wrought microstructure. This fact alone is likely to result in better mechanical properties of a

casting alloy as discussed next.

The presence of large silicon (Si) particles and acicular (needle-like) iron (Fe) intermetallic particles (highlighted by EDS maps in Fig. 7) in aluminum alloys adversely affects mechanical properties. Large Si particles act as stress concentrators, promoting crack initiation and reducing both ductility and fracture toughness due to their inability to deform plastically. They also contribute to non-uniform mechanical properties and increase tool wear during machining, leading to poor surface finishes. Similarly, acicular Fe intermetallics are brittle and serve as crack initiation sites, with their sharp morphology causing significant stress concentration. These particles hinder plastic deformation, reduce fracture toughness, and lower fatigue strength, while also being a common source of casting defects in alloys with high Fe content.

To mitigate these issues, alloy design and processing strategies such as refining Si particles with modifiers like strontium or sodium, reducing Fe content, and optimizing heat treatments can be employed. However, SolidStir® extrusion being a high strain rate thermomechanical processing technique, readily helps refine the particle morphology and distribution, thereby improving the mechanical performance of aluminum alloys. The severe plastic deformation and large processing strain results is very refined microstructure. The approach to use it for recycling is likely to be a game changer for the automobile and aerospace industry.

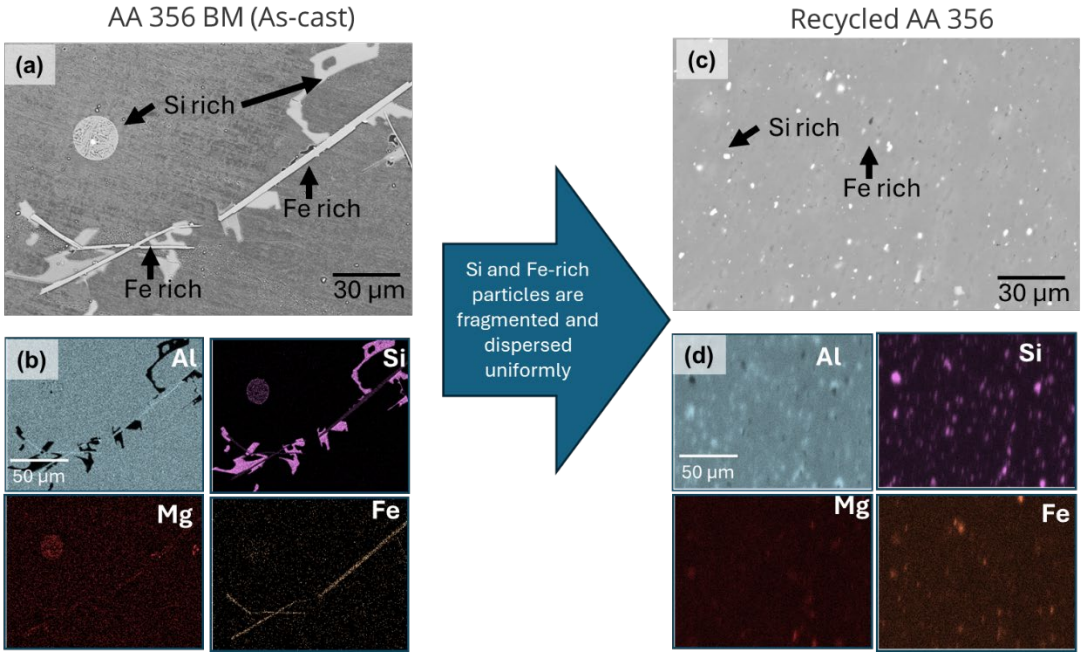

Figure 7: (a and c) SEM micrographs, (b and d) EDS maps of AA 356 BM and recycled AA 356 respectively.

EBSD micrographs were used to compare the microstructure of the recycled extrudate with the base materials (BM) for AA5083 and AA356 alloys. The AA5083 BM in the as-cast and T6 temper conditions exhibited an average grain size of 90.9±32 µm (Fig. 8(a)), while the AA356 BM showed a coarser average grain size of 150±100 µm (Fig. 8(b)). In contrast, the solid-state extruded (SSE) material underwent dynamic recrystallization during processing, resulting in a fine equiaxed microstructure (Fig. 8(c) and Fig. 8(d)). The recycled extrudate achieved an average grain size of 4.3±2.2 µm and 3.9±2.5µm for AA 356 and AA 5083, respectively, representing a reduction of more than twenty times compared to the BM. This significant grain refinement is expected to enhance the mechanical properties of the recycled extrudates through grain size (Hall-Petch) strengthening. Note that the strength (σ) is related to grain size (d) through the relationship, $\sigma = \sigma_0 + k\,(d)^{-1/2}$, where is σ_0 and k are constants. So, the current grain refinement adds significantly to grain boundary strengthening of the alloy.

The σ_0= 9.8 MPa is the frictional stress for Al, k = 90 MPa$\sqrt{\mu m}$ is the Hall–Petch coefficient, and d is the average grain size [19,20]. Considering the mean grain size (Fig. 7), $\sigma_{\text{Hall-Petch}}$ for BM is calculated as 19.2 MPa and 17.1 MPa for AA 5083 and AA 356 respectively. On recycling the grain boundary strengthening is enhanced for samples and

are calculated as 53.2 MPa and 55.3 MPa for the AA 356 and AA 5083, respectively. An enhancement of approximately 35 MPa in yield strength is attributed solely to grain boundary strengthening, achieved through the ability of the process to produce a refined equiaxed microstructure. This significant microstructural refinement, which is absent in melt-based recycling methods, highlights the superior capability of the solid-state recycling process.

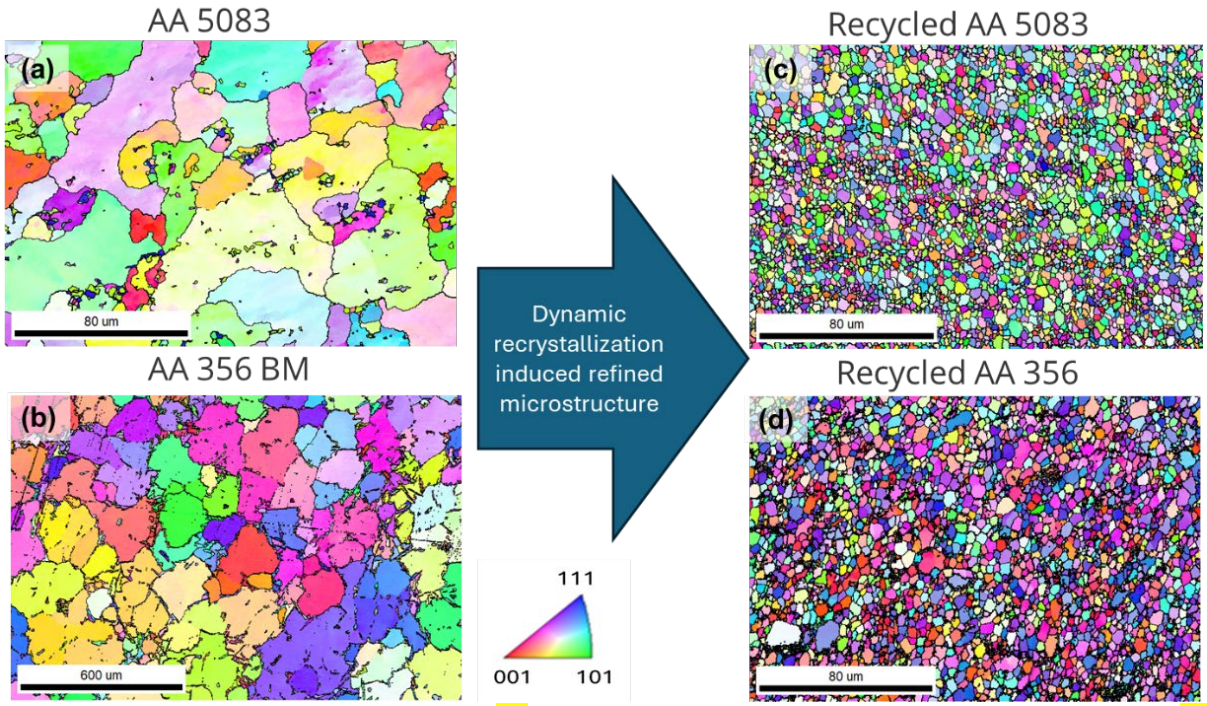

Figure 8: EBSD IPF map for (a) AA 5083 and (b) AA 356 base material, respectively, and SolidStir® recycled (c) AA 5083 and (d) AA 356, respectively.

4.2. Mechanical Properties

Figure 9 presents the tensile test results for the base materials (BM) and recycled materials. The AA356 BM exhibited a tensile yield strength (YS) of 125 MPa, an ultimate tensile strength (UTS) of 150 MPa, and a ductility of 2.5%. In comparison, the recycled material demonstrated a significant improvement, with the YS increasing to 225 MPa, the UTS rising to 300 MPa, and ductility increasing to 23%. For the AA5083 alloy, being a wrought alloy, the YS, UTS, and ductility remained consistent between the BM and the recycled extrudates. The recycled AA5083 alloy exhibited an YS of 200 MPa, a UTS of 320 MPa, and a ductility of 17%. Fractography of the recycled extrudates revealed the presence of dimples on the fracture surface, indicating ductile fracture behavior.

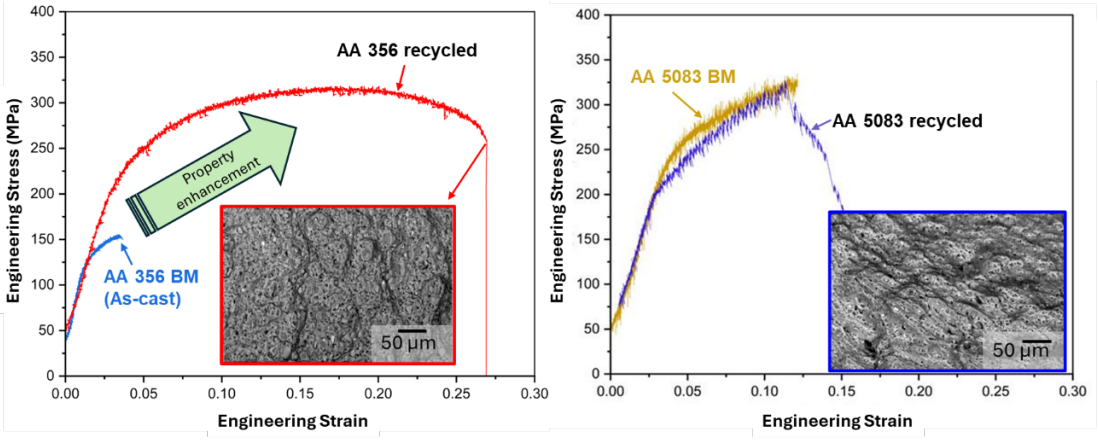

Figure 9: Tensile properties of SolidStir® recycled aluminum alloys with micrographs of fractured surface.

5. Conclusions and recommendations

This work highlights a new pathway for ***single step conversion from scrap to product without melting***. The innovative friction stir process based SolidStir® technology can revolutionize the recycling of aluminum scrap. With more than 3 billion pounds of aluminum available for recycling every year in US, converting a good fraction of that into upcycled aluminum alloy products will be transformative. As enabling technologies, SolidStir® represents an opportunity not previously possible. Demonstration of an enormous energy saving (>72%) due to solid-state processing and value addition (>10 fold) by converting low value metal scrap to high-valued components will gain immense attraction across various market segments [21]. Since aluminum usage is projected to increase in transportation industry for light weighting, the scrap metal generation during primary manufacturing and end of life components is expected to increase at a higher rate. Presently, due to the requirement of specialized furnace and casting equipment, automobile firms are selling the scrap to a third-party recycling firm for a cheaper rate per pound and buy the recycled and cast component at almost four times the price.

Aerospace companies are also interested in sustainability and recycling. The stringent requirements set forth for aerospace components demands looking into recycling technologies which enable strict control on alloy chemistry or can allow new ways for producing aluminum matrix composites with higher performance using scrap input. Leading aerospace companies are developing aircrafts for future with end-of-life recycling in mind and are also interested in technologies which can convert the 2XXX and 7XXX alloy machined chips with strict control over alloy chemistry after rigorous sorting. SolidStir® has the potential to address the concerns that these industries raised, capable of recycling the scrap with high purity. The economic benefit of current approach provides a natural driving force to embrace sustainability.

Figure 10 summarizes this new opportunity with including compositional aspects and shape/form aspects of the scrap and products. The new possibilities include incorporation of in-situ friction stir alloying and adding second phase particles for upcycling. For example, a 5XXX or 6XXX alloy can be converted to 2XXX or 7XXX alloy through in-situ alloying, thereby significantly enhancing the properties. Second phase particles like SiC and B_4C can be added to scrap to create aluminum matrix composite with enhanced fatigue and creep properties. Traditional powder metallurgy composite processing includes, atomization, mixing with second phase, consolidation and subsequent thermomechanical processing. These steps can be eliminated in the SolidStir® processing, and a single step can lead to extrudates or additively manufactured components.

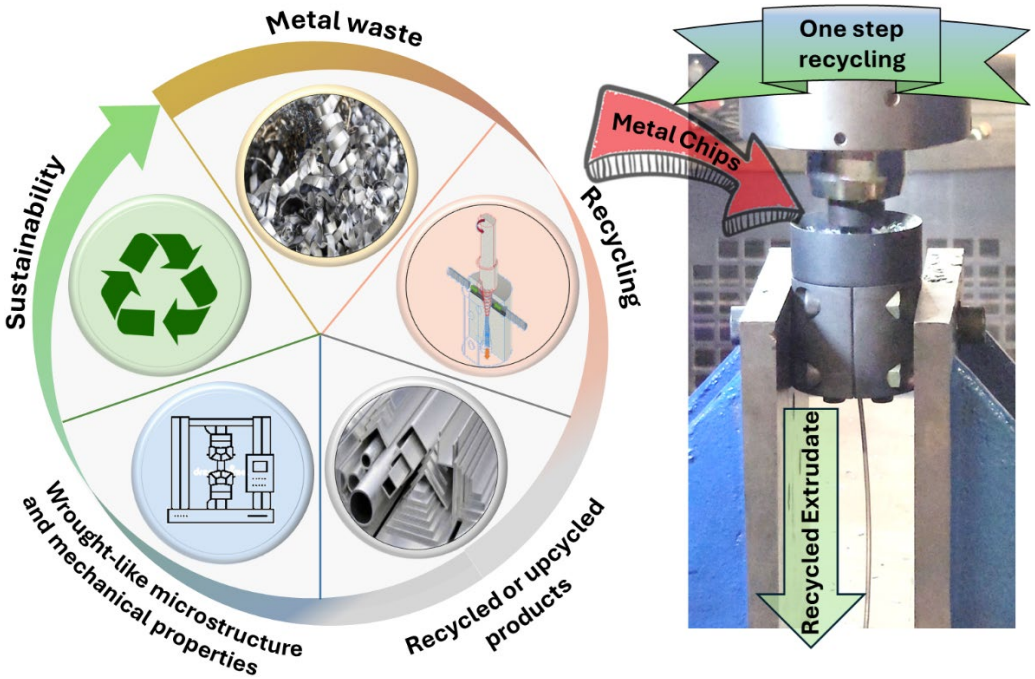

Figure 10: Redefining circular economy for metal recycling using solid-state SolidStir® technology as a step towards sustainable manufacturing.

6. Acknowledgement

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8. About the authors

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Professor Mishra is a leading authority in the field of friction-stir-derived technologies with more than 450 papers, h-index of 95 and more than 43000 citations. He has co-authored four books, 7 short books and has eight US patents.

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Dr. Haridas is one of the pioneer researchers of the proposed SolidStir® technology. He has been working on mechanical behavior of materials, structural mechanics, and solid-state processing over 10 years with 45 publications including 20 publications on friction stir-derived technologies.

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Dr. Gumaste's research focuses on improving structural performance through process-material co-design in friction-stir-based additive manufacturing. His work established foundational insights into SolidStir® technology. With 16 publications, 1 book chapter, and 1 patent, he continues to drive advancements in solid-state manufacturing.