Recycled aluminium feedstock in metal additive manufacturing: A state of the art review

Vladislav Yakubov\textsuperscript{a, **}, Halsey Ostergaard\textsuperscript{a,b}, Shishira Bhagavath\textsuperscript{c}, Chu Lun Alex Leung\textsuperscript{c,e}, James Hughes\textsuperscript{d}, Evren Yasa\textsuperscript{d}, Mani Khezri\textsuperscript{a}, Sandra K. L"oschke\textsuperscript{f}, Qing Li\textsuperscript{g}, Anna M. Paradowska\textsuperscript{a,b,g,*}

\textsuperscript{a} School of Civil Engineering, The University of Sydney, Sydney, NSW, Australia
\textsuperscript{b} Australian Nuclear Science and Technology Organisation, Kirrawee, NSW, Australia
\textsuperscript{c} Department of Mechanical Engineering, University College London, London, UK
\textsuperscript{d} University of Sheffield, Advanced Manufacturing Research Centre (AMRC), Sheffield, UK
\textsuperscript{e} The Research Complex at Harwell, Harwell Campus, Oxfordshire, UK
\textsuperscript{f} Sydney School of Architecture, Design and Planning, The University of Sydney, Sydney, NSW, Australia
\textsuperscript{g} School of Aerospace, Mechanical and Mechatronic Engineering, The University of Sydney, Sydney, NSW, Australia

\textbf{ARTICLE INFO}

\textbf{Keywords:}
Additive manufacturing
Recycled aluminium
Additive friction stir deposition
Environmental impact

\textbf{ABSTRACT}

Additive manufacturing has revolutionised the production of functional components and assemblies, offering a high degree of manufacturing flexibility. This review explores the latest advancements in additive manufacturing, focusing on its fusion-based and solid-state based technologies, and highlights the use of recycled aluminium as feedstock in these processes. The advantages and limitations of incorporating recycled materials are thoroughly analysed, considering factors such as material properties, sustainability, and process acceptance. While up to 14.4 kg CO\textsubscript{2} per kg of aluminium is released during primary aluminium ingot production, solid-state based additive manufacturing, which is tolerant of feedstock contamination, can directly recycle aluminium. Meanwhile, fusion based additive manufacturing can readily utilise recycling pathways such as maintaining grade, upcycling, and downcycling, as well as powder reuse, providing opportunities for significant emissions reduction. The examination of feedstock manufacturing in this review, such as wire for WAAM and powder for PBF, indicates that this step indirectly increases the resource consumption of additive manufacturing. Finally, the alignment of aluminium recycling and additive manufacturing with Circular Economy principles and the UN’s sustainable development goals are addressed, highlighting contributions to SDGs 3, 9, and 12.

1. Introduction

By offering geometric freedom and flexibility to manufacture one-off prototypes directly from a computer aided design, additive manufacturing (AM) of metallic materials allows product designers to push the boundaries of part performance. For example, additive
manufacturing has allowed weight reduction of automotive engine components [1], aerospace brackets [2], turbine engine components [3], and provided capability for inclusion of internal cooling channels or lattice structures not possible via conventional manufacturing processes [4]. AM is a disruptive technology as it reduces energy use, material waste, and maintenance costs, thus affecting the economic, societal, and environmental dimensions of sustainable development.

AM has been used commercially to reduce aviation’s carbon footprint through the use of topological optimisation. Najmon et al. [5] noted Airbus (France), Boeing (USA), General Electric (USA), and other major international aerospace companies have been applying AM to make topologically optimised components, such as nacelle brackets, fuel nozzles, and bleed pipes for enhancing efficiency of material usage, resulting in a high strength to weight ratio. Huang et al. [6] stated these components have saved up to 0.67 TJ of aviation fuel per aeroplane per year for every 100 kg weight reduction. In addition to topology optimisation, there are opportunities to further save material costs and improve manufacturing sustainability through the uses of feedstock recycling and next-generation AM technologies.

Considering the manufacturing process itself, the typical buy-to-fly ratio (a term that indicates the weight of purchased material to weight of finished part) reported by Kobryn et al. [7] for traditionally manufactured aircraft structures is 10:1 or higher whereas Rupp et al. [8] reported AM can improve this ratio to near 1:1.5. Spreafico and Landi [9] reported that such optimisation of the product structure is often proposed by both professional designers and students to achieve Circular Economy (CE) for products in general. Although AM offers many benefits, the material mining and refining processes for the production of AM feedstock are energy intensive and have a high carbon footprint. Norgate et al. [10] noted there is a need to address the challenges of recycling and reuse of metallic materials to reduce manufacturing’s environmental impact, which would also make AM more sustainable. However, Leung et al. [11] stated impurities in feedstock alter melt flow in fusion based process and as such has a complicated impact on defect formation.

One method to reduce material usage for the production of new feedstock is to direct-recycle waste materials generated by subtractive manufacturing processes. Such waste material is diverted from the traditional energy intensive recycling stream and hence no virgin material is used in the AM process.

For this review, an extensive literature search was conducted using Scopus, Web of Knowledge, and Google Scholar databases. The focus of the search was additive and traditional manufacturing methods and technologies and metal recycling. The following keyword examples were employed:

Additive manufacturing, AFSD, FSAM, WAAM, DED, LPBF, EPBF, Al recycling, direct recycling, recycling pathways, Al powder production, recycling friendly alloys, upcycling, downcycling, etc.

However, the source analysis is limited by the search language, which only encompassed articles in the English language, without any restrictions on subject area or publication date. Furthermore, there may be differences in methodology for reported CO₂ emissions and energy consumption figures between cited articles. Results were carefully reviewed and selected based on relevance and applicability to this review article.

The review is structured by first discussing the application of recycled and reused metallic material feedstock in AM. Then, different AM processes are separated based on their fundamental mechanisms, and the collecting, sorting, and processing steps are overviewed.

Fig. 1. Additive manufacturing process classification for common fusion based and solid-state based processes.
After this, a discussion is provided on defects, microstructure, and mechanical properties of parts manufactured using recycled materials. The energy savings and economic factors, as well as Sustainable Development Goals [12] are also considered. Furthermore, design for Circular Economy is addressed by identifying routes for utilising waste from subtractive manufacturing processes as feedstock for solid-state AM and exploring routes for reusing AM powder feedstock. By analysing a broad range of AM processes and addressing their unique requirements and current and developing recycling pathways, this review aims to provide a comprehensive understanding of the current state and future potential of sustainable additive manufacturing. For researchers who are new to the subject, the foundational information provided in this review offers an entry point to the topic. Meanwhile, this review offers a valuable resource to industry veterans and expert researchers in the field by providing a comprehensive update on the latest developments and considerations in sustainable fusion based and solid-state based additive manufacturing, allowing them to stay informed on advancements and potential areas for further research or implementation.

2. Additive manufacturing processes

AM is an umbrella term that describes the manufacturing of a part by material addition instead of casting or material removal. The American Society for Testing and Materials (ASTM) group ASTM F42 – Additive Manufacturing [13], developed a set of standards that divide AM processes into seven categories. This includes binder jetting (BJT), directed energy deposition (DED), material extrusion (MEX), material jetting (MJT), powder bed fusion (PBF), sheet lamination (SHL), and vat photopolymerization (VPP). Other additive processes exist but may not fit into ISO/ASTM 52900:2021, such as cold spray AM (CSAM), additive friction stir deposition (AFSD), and others. Nevertheless, common metallic AM processes can be classified based on their bonding method as shown in Fig. 1.

For the purpose of this review, metallic AM processes were divided into two main categories; (1) fusion based and (2) solid-state based (Fig. 1) according to the consolidation mechanisms.

2.1. Fusion based AM processes (liquid state)

Fusion AM includes laser and electron beam powder bed fusion (PBF) processes, which are laser powder bed fusion (LPBF, Fig. 2a) and electron beam powder bed fusion (EPBF). Furthermore, laser/electron/arc directed energy deposition (DED) and wire arc additive manufacturing (WAAM, Fig. 2b) are also fusion AM processes. Fundamentally, fusion AM applies a localised high energy heat source (laser, electron beam, or arc) to melt either wire or powder feedstock. Once the heat source is disarmed or moved away, the melt pool undergoes solidification, and subsequently phase transformation. Frazier [14] conducted a detailed review on each of the fusion processes.

The advantages of fusion processes include fine part resolution due to possibility of thin layer height (~30 μm) and small beam diameter (~50 μm), as well as the ability to manufacture highly intricate parts with small geometric features (~400 μm) [15].

However, Martin et al. [18] found the high thermal gradient induced by local melting causes solidification cracking in commercially important wrought alloy grades and leads to part distortion because of high residual stress. Furthermore, both feedstock and AM process are sensitive to oxygen contamination. Gheribi and Chartrand [19] noted increase in surface tension with oxygen content for liquid metals, reducing wettability. Yakubov et al. [20] found pores lined by oxides in LPBF-fabricated AlSi10Mg–4Ce containing oxidised Ce. Silva et al. [21] uncovered that aged AlSi10Mg powder did not appreciably differ in geometry from virgin powder, although sample fabricated from aged powder demonstrated 3.5 to 4.2 times greater porosity and 19.0% lower yield strength relative to virgin powder after DED fabrication. Leung et al. [11] found Marangoni convection changed from inward centrifugal to outward centripetal flow as a results of oxide presence for Invar 36 powder fabricated via laser additive manufacturing process. To ensure high melt wettability and good bonding of the solidifying material to the substrate, the feedstock materials and substrate surface should be free of oxides or other contaminants; and the related manufacturing processing should be conducted in a protective gas environment to

Fig. 2. Schematics of fusion AM processes showing: a) LPBF [16]; b) WAAM [17].
prevent melt oxidation and the formation of solidification defects, e.g. pores, cracks, and spatter during AM.

2.2. Solid-state based AM processes (deformation or plasticity based)

Solid-state based AM processes such as AFSD and FSAM fuse metal/alloys in the plastic state instead of using an external heat source to melt the feedstock materials. They have been applied for large structure manufacturing, for example an aluminium ring with diameter of 3.05 m as reported in Ref. [22]. Since manufacturing these large structures requires large amounts of aluminium, a large environmental benefit may be realised by using recycled material as feedstock. The foci for solid-state processes in this review are AFSD and FSAM, and the operating principles and consolidation mechanisms for AFSD and FSAM are similar to friction stir welding (FSW), where high pressure, high velocity, and friction are used to join feedstock consisting of metal sheets, powder, chips, and rods as discussed in Ref. [23]. Fig. 3a demonstrates FSAM, where multiple plates/sheets stacked on top of each other are welded together. Whereas AFSD in Fig. 3b uses rod form feedstock which adds onto the substrate material through a rotating motion; the material flows as a thick plasticised layer.

During solid-state processing, Mishra et al. [23] described the peak temperature at the workpiece as being lower than the alloy melting temperature (60–90% $T_m$). These parts do not suffer from solidification defects such as hot cracking or porosity noted for fusion processes such as LPBF [24], allowing AM of non-weldable Al alloy grades. Rivera et al. [25] applied AFSD to fabricate AA2219 with no defects and high mechanical properties. Mason et al. [26] fabricated AA7050 via AFSD and achieved a defect free structure, although hardness varied from initial layer to final layer. Solid-state AM presents several advantages for processing Al alloys such as the possibility to join dissimilar metals without forming a brittle interface and residual stress relatively lower compared to fusion AM processes due to the absence of high thermal gradients [27], and a high deposition rate allowing manufacturing of large components and assemblies [23]. However, the disadvantages of solid-state AM processes include coarse print resolution due to the large layer height and feedstock size, which requires post-machining to achieve the final geometry [27]. Processing of alloys with high hardness can cause tool wear during machining [23], and mechanical properties in as-manufactured condition may vary depending on location in structure [26].

2.3. Comparison between fusion based and solid-state based AM processes

As discussed in sections 3.1 and 3.2 and shown in Table 1, significant fundamental differences exist between fusion and solid-state AM processes, affecting aspects such as feedstock requirements, residual stress, and materials pallet. Therefore, selection of the appropriate processing method is dependent on the requirements of the final part and the material used.

In practical terms, solid-state AM processes are conducive to large structure fabrication due to high deposition rate. For example, Gopan et al. [28] reported an aluminium deposition rate of 1020 cm$^3$/h for AFSD (solid-state), which is much higher than 2.52–80 cm$^3$/h achieved by fusion processes. Meanwhile, Mishra et al. [23] stated approximately 11 kg/h and 10.5 kg/h deposition rate for AFSD and FSAM, respectively, and less than approximately 1.5 kg/h for DED and PFB processes. However, the high build rate comes at the cost of minimum feature size, which is 10 mm or larger for AFSD and submillimetre for PBF as reported in Ref. [27]. This limitation of AFSD is thought to be from tool design that prioritises high deposition rate, and it may be possible to achieve smaller feature size by reducing tool and feedstock size.

Residual stress is an important consideration for structural components since presence of tensile residual stress on surface promotes cracking, reducing fatigue life and resulting in early failure. Solid-state processes are well-known to result in low residual stress since no melting occurs and thermal gradient is lower compared to fusion processes [27].

The range of materials that can be processed is a significant distinction between solid-state and fusion processes. Solid-state processes offer more versatility and can fabricate wrought and casting grade Al alloys defect-free. In contrast, fusion processes suffer from solidification cracking and generally require eutectic or near-eutectic alloys [29], high temperature build preheating [30], or

Fig. 3. Schematics of solid-state AM processes showing: a) FSAM; b) AFSD [23].
grain nucleation particles [31] to achieve defect-free Al structures.

Contamination tolerance is another aspect important for defect-free structure fabrication and this topic is discussed in detail in section 5.1. Wan et al. [32] noted oxide on Al chip surfaces are broken during shear deformation, allowing for direct contact between metal surfaces and bonding during deformation processing. Yoder et al. [33] examined as-received Al machining chips and AFSD structure fabricated from these chips and noted breakup of oxides layers and Fe-containing brittle intermetallics. Fusion processes on the other hand are sensitive to feedstock impurities and presence of oxygen in processing environment, requiring feedstock handling and storage considerations as well as and an inertised build chamber or shielding gas as discussed in section 2.1. Furthermore, feedstock geometry differs between solid-state and fusion processes. Whereas solid-state processes use chips, powder, sheets, or bars, fusion processes require feedstock in the form of powder or wire.

<table>
<thead>
<tr>
<th>Attribute</th>
<th>Solid-state</th>
<th>Fusion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Component size</td>
<td>Large</td>
<td>Small</td>
</tr>
<tr>
<td>Minimum feature size</td>
<td>~10 mm [27]</td>
<td>&lt;1 mm [27]</td>
</tr>
<tr>
<td>Residual stress</td>
<td>Low</td>
<td>High</td>
</tr>
<tr>
<td>Materials pallet</td>
<td>Broad</td>
<td>Limited</td>
</tr>
<tr>
<td>Contamination tolerance</td>
<td>Good</td>
<td>Poor</td>
</tr>
<tr>
<td>Feedstock geometry</td>
<td>Sheet, powder, chips, rods</td>
<td>Powder, wire</td>
</tr>
<tr>
<td>Processing environment</td>
<td>In air</td>
<td>Inert (Ar, N)</td>
</tr>
</tbody>
</table>

Table 1
Overview of solid-state and fusion AM process attributes.

Fig. 4. Flow cycle of Al recycling processes and AM technology material acceptance for a) direct-recycling; b) maintaining grade, upcycling, and downcycling. Graphs illustrating relative attributes for c) casting, solid-state, and fusion processes; d) AM processes.
3. Recycled materials

Aluminium recycling provides an opportunity for reduction of greenhouse gas emissions since it reduces the need for primary aluminium production, which has been identified in critical lifecycle assessment by Paraskeva et al. [34] as an energy- and carbon-intensive process. Al alloys are used in an extremely wide range of applications and industries, including aerospace, automotive, construction, and beverage packaging, all of which generates a large quantity of waste. Therefore, solid-state AM processes could be a key enabler for the large-scale Al recycling.

Spreafico and Landi [9] analysed the use of design strategies to implement CE in products by collecting and analysing data from students and professional designers. The authors reported that the recycling of the material of the product was a popular approach for both students and professionals, although professional designers noted various challenges in implementation of this strategy.

Additionally, Raabe et al. [35] have reported the current AI waste stream as being underutilised due to challenges in separation of mixed alloy stream, accumulation of impurity elements, and removal of impurities. Even though impurity removal can be performed through slag phase oxidation or gas phase evaporation, the recycled Al is often diluted with primary Al to achieve standard compositions, e.g. Al6061, A356, etc. However, compositionally closed recycling loops and development of mixed or recycling friendly alloy grades have been explored in Ref. [36]. In the case of AM, the recycling process depends on the waste stream quality and final product requirements. To date, direct-recycling (melt-free) of Al feedstock via AM has only been performed through solid-state AM processes (Fig. 4a). Meanwhile, maintaining grade and up/downcycling have been applied for both fusion and solid-state processes (Fig. 4b). Fig. 4c and d compare the different relative attributes of solid-state, fusion, and casting processes.

3.1. Traditional recycling pathways: upcycling, maintaining grade, and downcycling

As discussed in section 4, it is difficult to remove impurities or unwanted alloying elements from Al waste stream, which usually consists of unsorted Al alloy grades. However, Al producers have developed cost effective methods to utilise waste stream Al as a minor addition or major component of their products. These methods include upcycling, maintaining grade, and downcycling, and process selection depends on economic factors, input waste stream composition, and target product composition.

For the case of upcycling, the melt consisting of recycled Al is diluted with a high purity primary Al because the recycled wrought alloy grade has a lower alloying element content and a lower tolerance for impurities compared to the casting grades of Al as discussed in Ref. [35].

A compositionally closed recycling loop ensures the input material has the same composition as the target Al alloy (maintaining grade), eliminating dilution with primary Al. This method results in a low virgin material usage and hence lowers the carbon emissions of the process whilst fulfilling end product requirements. This has been applied in a small scale and various closed-loop recycling operations in the manufacturing sector such as the beverage packaging [37] and automotive industries [38], wherein a fixed aluminium alloy grade is selected. At a larger recycling scale, nevertheless this becomes impractical as it is difficult to separate various alloy grades from the recycling waste stream.

For the case of downcycling, wrought grades are used as an input to make lower purity casting grades. Cast alloy grades have wider compositional limits and a higher alloying element content relative to wrought grades, e.g. the A380 and Al6061 alloys contain 86.7 wt % and 97.9 wt% of Al, respectively as mentioned in Ref. [39]. The wrought alloy grades can be melted and alloying elements added to achieve target composition with little or no primary aluminium addition. Raabe et al. [35] noted this method is widely utilised since much of the Al in an unsorted waste stream consists of wrought grades, making downcycling a cost-efficient process for industry.

3.2. Considerations for fusion based processes

As discussed in section 2.3, fusion based processes are sensitive to material composition and suffer from solidification cracking. The layer-by-layer melting and solidification process requires high surface wettability to ensure good layer bonding and minimise porosity formation during AM [20], and even a minor change in material composition requires considerable changes in processing parameters to achieve target mechanical properties [21]. The high surface area of powder feedstock leads to rapid pickup of oxygen, forming both oxides and hydroxides, adventitious carbon, and moisture during powder handling and storage [40]. However, while a review by Mighmian et al. [41] found that surface oxides causes porosity, poor bonding, and spatter, Leung et al. [11] found that pores induced by oxides may be released via keyhole melting and the Marangoni flow inversion.

Even though downcycling and compositionally closed recycling loops are attractive for recycled feedstock manufacturing, other recycling opportunities exist in the case of powder feedstock. Most of the unconsolidated powder retains its shapes and composition while surrounding the manufactured parts despite heat source interaction with powder bed leading to some partially melted particles and spatter. Cordova et al. [42] reported blending used feedstock with virgin powder and Weiss et al. [43] implemented simple sieving operations prior to powder reuse. In both instances it was possible to reuse powder several times while maintaining acceptable manufactured part quality; however, change in mechanical properties after a certain number of reuse cycles was noted.

3.3. Considerations for solid-state based AM processes

As discussed in section 2.2, solid-state AM processes have been successfully manufacturing a wide range of Al grades. They can tolerate minor contamination such as elevated Fe content or surface oxidation, though some degree of contamination control is necessary to achieve high mechanical properties and low defect level [33]. They use upcycling, compositionally closed loops, and
downcycling as recycled feedstock materials in a form of powders, chips, and rods. Furthermore, Yoder et al. [33] applied AFSD to direct-recycle cast Al chips from automotive industry, achieving higher ductility than bulk material. Similarly, Babaniaris et al. [44] used AFSD to direct-recycle AA6063 and noted reduction in size of harmful Fe-containing intermetallics. The resource-efficient direct-recycling (melt-free) of waste from subtractive manufacturing or other processes generally requires lower energy input and results in less material loss compared to traditional recycling processes involving remelting.

3.4. AM-specific recycling pathways: direct-recycling and powder reuse

3.4.1. Direct-recycling

Direct-recycling offers a more sustainable and cost-effective approach to AM compared to conventional remelting methods. Using waste from subtractive manufacturing processes or the recycling stream as feedstock for solid-state AM, the discarded materials may become value added, maximising the usage of abundant resources, minimising waste, and lowering the overall energy required to produce a part.

Raabe et al. [35] noted traditional remelting recycling poses several problems including (1) high metallic material loss due to the presence of surface oxides, (2) generation of toxic gas from combustion of surface contaminants such as oil, and (3) high energy consumption of using hot furnaces to remelt materials. In contrast, Wan et al. [32] stated that direct-recycling, a melt-free process relying on solid-state deformation, mitigates the aforementioned problems.

Solid-state AM processes can manufacture parts using all types of Al alloy feedstocks, this minimises the need for sorting out Al waste stream. They can be used for construction of civil structures, consumer products, and defence applications which enable utilisation of 3% and 35% for honeycomb structure and a gear, respectively. The gas atomised powders being used in PBF and DED are expensive as they are made by an energy intensive production process. Increasing powder reuse and recycling via solid-state processes creates an outlet for scrap powder, lowering the cost to the value chain and environmental impact associated with AM. The effect is more pronounced with expensive materials, e.g. Ti and Ni-based alloys.

Due to the accumulation of oxides/hydroxides, carbon, and other impurities as well as dimensional changes caused by partial melting and storage, extensive powder reuse will eventually result in AM parts manufactured with undesirable mechanical properties or non-conformance with material composition specifications as demonstrated in powder reusability study by Santecchia et al. [49]. To combat this issue, several powder reuse strategies are discussed by powder and AM machine manufacturer General Electric [50]. These strategies include the 2-bin method, where powder accumulated in overflow bottle is sieved, labelled with the total number of use cycles, and then reused in the main powder supply up to the reuse limit; the virgin blend method, where the powder accumulated in overflow bottle is sieved and blended with virgin powder, labelled with the total number of use cycles, and then reused in the main powder supply up to the reuse limit; and the top-up newest method, which is a hybrid approach of the 2-bin and virgin blend methods where any powder with lower reuse cycles can be blended with overflow bottle powder instead of just virgin powder.

Powder reuse is an ongoing research topic, e.g. Denti et al. [51] explored average usage time (AUT) as a more reliable method for determining the powder replacement or refreshing interval. In addition, Rojas-Diaz et al. [52] applied powder metallurgy (PM) techniques to manufacture samples from mechanical saw chips. Similarly, Parucker and Costa [53] applied PM to fabricate samples from grey cast iron swarf. It is thus envisioned that PM may also be a recycling outlet for AM powder.

4. Part manufacturing

AM processes are rather complex, and material properties depend strongly on their processing parameters and history [54]. Traditionally, it has been difficult to maintain reproducibility and repeatability of part performance, resulting in variations in mechanical properties between builds [55]. To remove sources of uncertainty, feedstock with low impurities content and no composition variation have mainly been used for AM in industry, particularly for production of components to be used in safety critical applications. This section reviews the formation and evolution of microstructure and the resultant mechanical properties of part made using recycled feedstock.

4.1. Oxide-induced defects

The presence of oxides generally increases the frequency of defects, although the pathway is not straightforward as discussed in Ref. [11]. Srivastava and Meshram [56] noted that during traditional recycling (melt-based), oxides can be removed. However, oxides are not removed during solid-state manufacturing. Jerina et al. [57] stated that large surface area favours relatively greater environmental exposure-induced oxide. Therefore, due to their large surface area, powder, swarf and chip reuse and direct-recycling routes may introduce elevated oxide levels into manufactured structures.
Since fusion processes result in melt formation, low surface energy at aluminium melt and oxide interface causes low melt wettability, subsequently leading to poor layer bonding [58]. Whilst swarf and chips have been used to achieve powder via ball milling [59] or wire via friction stir extrusion (FSE) [60], swarf and chips may present high oxide content due to their large surface areas [57]. Therefore, care must be taken when selecting the feedstock source since presence of oxides creates favourable conditions for porosity. Low oxide wettability is known to cause lack of fusion (LoF) porosity during LPBF [61]. In laser-DED (L-DED) [21] and arc-DED [62], oxides and hydroxides present on particle surface, melt pool, or wire contribute to gas porosity via hydrogen inclusion. However, in the case of arc-DED, positive ion bombardment via alternating current instead of direct current effectively removes oxides and reduces porosity, providing a potential route for FSE-recycled feedstock wire [63].

Since no melting occurs in solid-state AM processes in which the material temperature is well below the melting point, oxide-induced defects described for fusion processing of Al are not relevant. During friction stirring, the oxides are broken up due to shear deformation, which allows for clean surface contact [32]. Plastic deformation at elevated temperature results in high strength atom diffusion bonding. The nodular oxides are dispersed through the Al matrix though they do not induce embrittlement [33]. Nevertheless, the surface oxidation favours the formation of kissing bonds at layer or plate interfaces, in which case two surfaces are in contact with each other and appear to be strongly bonded but are interfaced by an oxide layer [64]. This oxide layer causes the bond to have little to no strength. Stronger stirring to control kissing bonds can be achieved via a higher application pressure, increased heat, higher tool speed, and increased plunge depth.

4.2. Microstructure development and mechanical properties

For fusion processes, the microstructure development is significantly influenced by the cooling rate which is affected by processing parameters, e.g. laser power, spot size, scan speed, etc [65]. However, the resultant microstructure is inherently different to casting, owing to the high thermal gradient and fast-moving solidification front. Casting can only achieve high cooling rate for small or thin sections, such as filaments or ribbons [66] whereas fusion AM uses the heat source to melt small local volumes, implying the underlying material functions as a heat sink allowing cooling rates up to $10^7$ K/s in AM comparison to 10 K/s in casting [67]. Rapid solidification favours the development of a fine microstructure and enables α-Al alloying element supersaturation and corresponding solid-solution strengthening effect, allowing for higher mechanical properties in the as-printed condition than that achieved via solid-state processes. However, directional grain growth often occurs, resulting in anisotropic mechanical properties for fusion-based AM [68].

For solid-state AM processes, material joining relies on frictional heat, high strain rate, and material flow [23]. These processes usually form a homogeneous recrystallised microstructure consisting of refined equiaxed grains, increasing the strength of non-age

Table 2
Mechanical properties comparison for various aluminium alloys, manufacturing technologies, and post processing steps.

<table>
<thead>
<tr>
<th>Material</th>
<th>Process</th>
<th>Treatment</th>
<th>Yield (MPa)</th>
<th>UTS (MPa)</th>
<th>Elongation to fracture (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AlSi10Mg</td>
<td>LPBF [43]</td>
<td>None</td>
<td>220</td>
<td>369</td>
<td>8.9</td>
</tr>
<tr>
<td></td>
<td>EPBF [80]</td>
<td>None</td>
<td>62</td>
<td>137</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td></td>
<td>T6</td>
<td>249</td>
<td>305</td>
<td>17.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>LDED [81]</td>
<td>None</td>
<td>200</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>None</td>
<td>107</td>
<td>209</td>
<td>6.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cast [52]</td>
<td>Aging</td>
<td>N/A</td>
<td>300–317</td>
</tr>
<tr>
<td></td>
<td></td>
<td>High pressure die cast [82]</td>
<td>None</td>
<td>N/A</td>
<td>300–350</td>
</tr>
<tr>
<td></td>
<td></td>
<td>High pressure die cast [82]</td>
<td>T6</td>
<td>N/A</td>
<td>330–365</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>T6</td>
<td>288</td>
<td>323</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cast [70]</td>
<td>T6</td>
<td>106</td>
<td>162</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Hot forged [84]</td>
<td>None</td>
<td>411</td>
<td>431</td>
</tr>
<tr>
<td></td>
<td></td>
<td>T6</td>
<td>308</td>
<td>340</td>
<td>11.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cast [70]</td>
<td>T6</td>
<td>109</td>
<td>225</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Hot forged [84]</td>
<td>None</td>
<td>452</td>
<td>518</td>
</tr>
<tr>
<td></td>
<td></td>
<td>T6</td>
<td>156</td>
<td>339</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td></td>
<td>T6</td>
<td>507</td>
<td>566</td>
<td>9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>T6</td>
<td>472</td>
<td>550</td>
<td>4.6</td>
</tr>
<tr>
<td>AlSi10Mg powder 1x use</td>
<td>LPBF [43]</td>
<td>None</td>
<td>230</td>
<td>398</td>
<td>9.7</td>
</tr>
<tr>
<td>AlSi10Mg powder 1x use</td>
<td>LPBF [77]</td>
<td>None</td>
<td>172</td>
<td>270</td>
<td>7.1</td>
</tr>
<tr>
<td>AlSi10Mg powder 1x use and 96h aged</td>
<td>LPBF [77]</td>
<td>None</td>
<td>150</td>
<td>229</td>
<td>3.4</td>
</tr>
<tr>
<td>AlSi10Mg powder 96h aged</td>
<td>LDED [21]</td>
<td>None</td>
<td>87</td>
<td>178</td>
<td>13.6</td>
</tr>
<tr>
<td>AlSi083-direct recycled</td>
<td>AFSD [86]</td>
<td>None</td>
<td>179</td>
<td>368</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>Cast [86]</td>
<td>H131</td>
<td>286</td>
<td>343</td>
<td>14</td>
</tr>
<tr>
<td>A3xx-direct recycled</td>
<td>AFSD [53]</td>
<td>None</td>
<td>193</td>
<td>173–200</td>
<td>&lt;1</td>
</tr>
<tr>
<td></td>
<td>Cast [33]</td>
<td>N/A</td>
<td>175–200</td>
<td>17.8</td>
<td></td>
</tr>
</tbody>
</table>

V. Yakubov et al.
hardenable alloys (1000, 3000, and 5000 series) [69], but causing undesirable precipitation and hardness gradients in age-hardenable alloys (2000, 6000, and 7000 series) due to thermal history [25]. Table 2 shows that AFSD of age-hardenable alloys (Al6061 and Al7075) results in a higher ductility and a lower yield strength compared to the as-cast material, though AFSD-manufactured parts with T6 heat treatment increases strength and reduces ductility via precipitate hardening [70].

The main problem of Al recycling and reuse for AM is the accumulation of impurities, particularly Fe and O. Fe has high solubility in Al melt and forms brittle intermetallics during solidification [71], inducing negative effect on mechanical properties for castings [72]. For fusion AM processes, the impact of Fe contamination has not yet been well explored; however, Fe contamination has been demonstrated to form needle-like β-Fe intermetallic phases in LPBF-manufactured AlSi10Mg [73]. In the case of arc-DED, needle-like Fe-rich intermetallics were observed throughout the as-deposited Al6061 microstructure but these needles became smaller during T6 heat treatment as seen in Fig. 5a and b [74].

Considering the impact of oxygen contamination during powder reuse, careful handling and storage can allow inconsequential oxygen increase and little impact on mechanical properties over several reuse cycles [43]; however, factors such as powder size and storage environment affect oxidation rate [75], and AlSi10Mg powder oxygen content has been demonstrated to double after six reuse cycles [76]. Fedina et al. [77] noted a direct correlation between powder oxygen content, sample porosity, and mechanical properties for LPBF-manufactured AlSi10Mg, with 0.072 wt% O content resulting in 0.97% sample porosity, an ultimate tensile strength (UTS) of 269.7 MPa, and 7.1% elongation to fracture, while 0.274 wt% O content resulted in 10.1% sample porosity, UTS of 229 MPa, and 3.4% elongation to fracture. Similar trends have been noted for other fusion processes and for other materials, highlighting the high oxygen sensitivity [21,41,78]. An example of a pore caused by oxide inclusion can be seen in Fig. 5c.

Solid-state AM have demonstrated relatively high tolerance for oxides and impurities. This is because shearing during solid-state manufacturing breaks up oxides and needle-like intermetallic precipitates, resulting in uniform distribution of fine particles relative to feedstock as demonstrated in Fig. 6a–c. Furthermore, the difference of elastic moduli between the matrix and these particles is thought to result in strain hardening, contributing to increase in UTS and yield strength. Babaniaris et al. [44] reported the average intermetallic particle size for extruded direct-recycled Al6063 feedstock to be $3.47 \pm 2.38 \mu m$, which was reduced to $1.08 \pm 0.75 \mu m$ after AFSD. Yoder et al. [33] used mixed scrap Teksid Al alloys (comparable to 3xx cast alloy) as direct-recycled AFSD feedstock and achieved an UTS of 193 MPa and an impressive 17.8% elongation to fracture in the as-deposited condition. In comparison, this feedstock material in as-cast condition achieved 175–200 MPa UTS and <1% elongation to fracture.

![Fig. 5. Fe intermetallics (white phase) in a) as-deposited and b) T6 heat treated arc-DED Al6061 [74]. c) Oxide-induced pore in LPBF-manufactured AlSi10Mg [61].](image-url)
5. Energy requirements and resource usage

The advantages of AM over subtractive manufacturing processes are: reduced material waste and the ability to manufacture lightweight, complex geometry, and near-net shaped parts. Less input material signifies a key feature for sustainable AM. Even though this advantage is strongest when primary Al is used for feedstock production, overall greenhouse gas emissions are still lower using recycled Al feedstock.

A comprehensive lifecycle analysis revealed that primary Al ingot production releases up to 14.4 kg CO$_2$ per kg Al when assessed based on global average production emissions, and this value is reduced to 3.5 kg CO$_2$ per kg Al when assessed based on the best available production technology [87]. Meanwhile, the CO$_2$ emissions are reduced to 0.6 kg per kg Al for ingot production via recycling [88]. Fig. 6 shows that the energy demand for raw material production, AM materials manufacturing, and product manufacturing highly depend on the selected materials. For example, average ingot alloy production for a titanium aerospace part requires substantially higher energy in comparison to an average Al ingot production [89]. Additionally, the entire life cycle analysis of AM parts needs to be taken into account for a more accurate comparison since it is not fully reasonable to compare two different manufacturing routes where AM can produce parts that are impossible to make otherwise. In the case of titanium aerospace part production, the weight advantage of AM processes leads to significant energy savings during the product use phase and this needs to be taken into account in future studies.

The studies focus on the recycling of material for AM applications are limited and there is a gap in literature regarding energy requirements. Table 3 shows AM requires additional energy input to process ingots into useable feedstock, and this is typically not necessary for traditional processes such as casting. The total energy input for AM is therefore influenced by the recycled material usage, feedstock geometry, and the processing steps required to prepare the material for AM [89].

<table>
<thead>
<tr>
<th>Part and production method</th>
<th>Ingot production (kWh/kg)</th>
<th>Powder atomisation (kWh/kg)</th>
<th>Manufacturing, no post processing (kWh/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal structure, LPBF</td>
<td>100–400</td>
<td>10–30</td>
<td>50–100</td>
</tr>
<tr>
<td>Al structure, die cast</td>
<td>50–100</td>
<td>0</td>
<td>&lt;10</td>
</tr>
<tr>
<td>Ti aerospace structure, machined</td>
<td>300–400</td>
<td>0</td>
<td>&lt;10</td>
</tr>
<tr>
<td>Turbine blade, cast and machined</td>
<td>50–100</td>
<td>0</td>
<td>&lt;20</td>
</tr>
</tbody>
</table>

Fig. 6. Fe intermetallics and Si in direct-recycled mixed Teksid Al-alloy a) extruded feedstock bar and b) as-deposited AFSD [33]. c) Oxides present at layer interface in AFSD as-deposited Al6061 [79].
5.1. Gas atomised powder

Powder materials may require the highest additional energy input of all discussed forms. Gas atomisation is a common approach used for producing Al alloy powder feedstock for AM, which has modest production yield and high resource requirements. During gas atomisation, material is heated to melting point in a tundish by an induction coil. This melt exits an orifice and is struck with a high pressure inert gas flow to facilitate atomisation, see detailed review of various powder production process in Ref. [90].

Although the particle size distribution is controllable by changing process parameters such as orifice diameter, not all particles fall into the size range suitable for AM (23–45 μm for LPBF) and (70–105 μm for DED). Marinucci et al. [91] used a commercial gas atomiser to manufacture AlSi10Mg powder from melt, and after sieving to size range of 20–50 μm, obtained 30% powder fraction relative to original powder amount. With innovative nozzle design and process optimisation, the desired particle size fraction can be increased and rejected particles can be remelted [92].

Another factor impacting the input energy requirements for gas atomisation is the purity and velocity of inert gas. The gas selection is conducted based on thermal conductivity, reactivity with the melt, and cost. For Al alloys, nitrogen is commonly used, although argon and helium are also suitable. Furthermore, the particle size generally decreases with increasing gas/metal flow ratio (kg/kg). Ünal [93] reported that a gas pressure of 1.05 MPa with gas/metal flow ratio of 4 generates 52 μm mean particle diameter, while gas pressure of 1.56 MPa with gas/metal flow ratio of 8 generates 27 μm mean particle diameter.

The European Industrial Gas Association (EIGA) reported that 99.9% purity liquid nitrogen produced through an air separation process consumes ~0.55 kWh of energy per kg of nitrogen [94]. Therefore, 1 kg of gas atomised Al alloy powder would require a gas/metal flow ratio of 6, equivalent to 3.3 kWh of energy to produce the nitrogen gas used in the process. This is estimate assumes that all produced powders are used in the process and does not consider the production yield.

Fig. 7. a) Benefits of AM linked to SDGs; b) Process steps in closed lifecycle AM.
5.2. Wire consumable

Wire feedstock used for arc-DED processes today are usually ordinary welding wire, though specialty AM wire has recently emerged on the market [95]. The manufacturing process for Al welding wire typically involves continuous casting followed by drawing [96]. However, extrusion has also been demonstrated to be suitable [97]. Baffari et al. [98] investigated energy and material usage for several different manufacturing processes for fabricating wire from waste AA2050 chips. It was found that the casting and drawing process consumed 12.45 kWh per kg of wire produced if permanent material loss due to dross formation was not considered [99]. However, permanent material loss of 15% due to dross from melting waste AA2050 chips increased overall energy consumption of the process. Considering material loss, an energy requirement of 28.64 kWh per kg wire produced was calculated. In the same study, equal channel angular pressing-hot extrusion and friction stir extrusion did not result in material loss, in which required 13.89 and 9.60 kWh per kg wire produced, respectively.

6. Opportunities, challenges, and conclusions

The recycling of aluminium feedstock in metal additive manufacturing is well aligned with Circular Economy principles which support the reuse and regeneration of materials and products as part of a no-waste economy, and the replacement of traditional manufacturing solutions with sustainable advanced technology [100]. The UN has recognised the transition to CE as a vital milestone in achieving the UN climate goals by 2050 [101], and the wider use of recycled aluminium as feedstock in additive manufacturing may represent a significant stride towards achieving multiple Sustainable Development Goals (SDGs), as shown in Fig. 7a. For example, Goal 3, ensuring good health and well-being, is intertwined with the reduction of greenhouse gas emissions and environmental pollution through decreased energy-intensive primary aluminium production. Moreover, Goal 9 and 12, which focuses on industry, innovation, and infrastructure, is achieved since convergence of recycling with additive manufacturing fosters technological advancement, encouraging the growth of cleaner and more efficient industrial processes and stimulating economic progress.

As Australia’s largest metal manufacturing companies need to meet their mandatory climate-related reporting targets under the Green House Gas (GHG) reporting standards, a switch to fusion based and solid-state based technologies represents opportunities for effective decarbonisation because of their lower energy requirements and waste-based resource usage as discussed in section 5. The GHG protocol includes direct emissions from owned or controlled sources (scope 1) and indirect emissions in the value chain (upstream and downstream), such as purchased goods, waste from operations; processing & use of products (scope 3) that will be mandatorily reported from 2024 onward [102].

Government policies play a pivotal role in assisting the transition to CE for aluminium alloys manufacturing. In addition to mandatory reporting schemes targeting the largest companies only, government interventions could be extended to address the entire sector and provide incentives to smaller manufacturers to adopt closed-loop lifecycle systems (Fig. 7b), enabling the recovery of valuable resources from end-of-life products. Creating frameworks for eco-design and promoting extended producer responsibility would ensure products are designed for durability, repairability, and recyclability. AM’s importance for effective sustainable production is underscored by its wide and diverse range of applications, spanning aerospace, automotive, construction, and industries that would benefit from the large-scale implementation AM.

However, despite the environmental and economic advantages of aluminium recycling, its full potential remains untapped due to technical challenges in separating mixed alloy streams and impurity element accumulation. In the realm of AM, the applicability of the recycling process relies on waste stream quality, AM technology, and final product requirements. Solid-state AM processes have been employed for highly efficient and environmentally friendly direct-recycling thanks to the high oxide tolerance and melt-free processing, making it possible to reuse mixed aluminium chips from subtractive manufacturing processes as feedstock. Meanwhile, both fusion and solid-state methods may use up/downcycled and maintaining grade recycled aluminium feedstock, provided the composition meets the requirements for the alloy grade. Nevertheless, additional energy input is required to achieve the required feedstock geometry for AM, and this topic has not been well-explored. Additionally, there is a limited body of research focusing on the recycling of materials for AM applications, creating a gap in the literature when it comes to addressing the performance and quality of AM components produced from recycled materials. Bridging this gap through further studies will be vital for unlocking the full potential of recycled materials in AM and sustainable manufacturing practices.

Funding

Vladislav Yakubov acknowledges financial support from Solid-State Additive Manufacturing for Recycled Aluminium Alloys, DVC Research/Sustainable Development Goals collaboration program (G217531).

Data availability statement

This study adopts a systematic review approach, including all relevant data within the manuscript.

Ethics statement

An ethics statement is not required as this study solely relies on previously published literature.
CRediT authorship contribution statement

Vladislav Yakubov: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. Halsey Ostergaard: Writing – review & editing, Writing – original draft, Formal analysis, Conceptualization. Shishira Bhagavath: Writing – review & editing, Writing – original draft, Resources, Project administration, Funding acquisition, Conceptualization. Chu Lun Alex Leung: Writing – review & editing, Writing – original draft, Resources, Project administration, Funding acquisition, Conceptualization. James Hughes: Resources, Project administration, Funding acquisition, Conceptualization. Even Yasa: Writing – review & editing, Writing – original draft, Resources, Funding acquisition, Conceptualization. Mani Khezri: Writing – review & editing, Methodology, Formal analysis. Sandra K. Loschbe: Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition. Qing Li: Supervision, Resources, Project administration, Funding acquisition, Conceptualization. Anna M. Paradowska: Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors acknowledge the support from the Research Complex at Harwell (RCoA), funded through the UK-EPSRC MAPP Future Manufacturing Hub (EP/P006566/1), Manufacturing by Design (EP/W003333/1), Made Smarter Innovation - Materials Made Smarter Research Centre (EP/V061798/1); Data-driven, Reliable, and Effective Additive Manufacturing using multi-BEAM technologies (EP/W037443/1), and performance-driven design of aluminium alloys for additive manufacturing (PAAM) (EEP/W006774/1).

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

