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RESEARCH ARTICLE

How Cerium and Lanthanum as Coproducts Promote Stable Rare Earth Production and New Alloys

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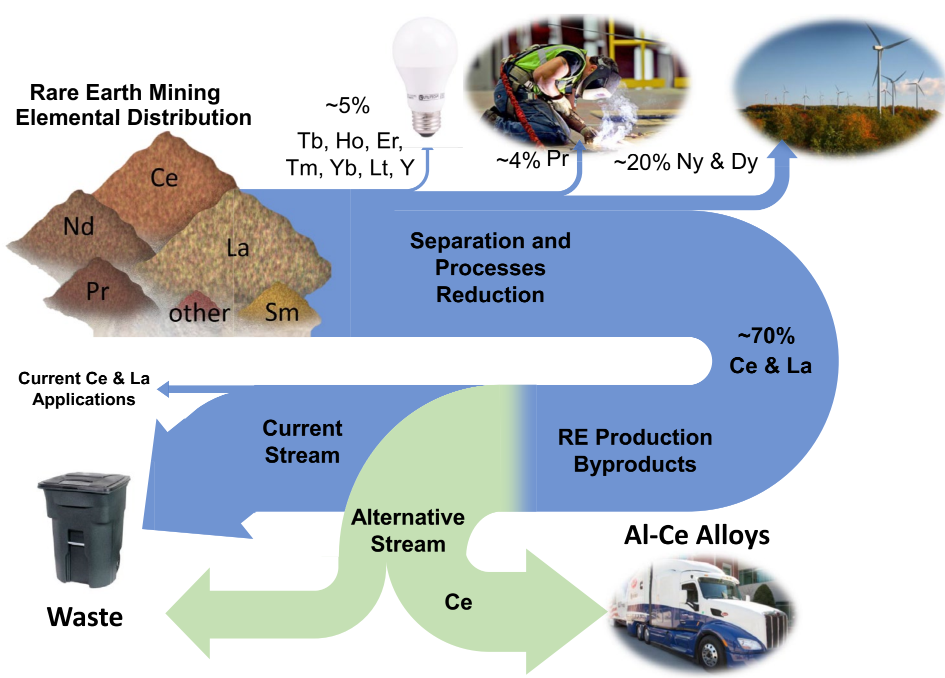
Abstract
The largest outputs of rare earth mining are the low-value byproducts cerium and lanthanum, which burden rare earth supply chains because they must be separated from more desirable rare earths used in magnet production. Promoting demand for cerium and lanthanum can potentially diversify the economics of rare earth mining and improve supply chain stability for all rare earth elements. A promising avenue for increasing byproduct rare earth element demand is their use in aluminum alloys; an application for cerium and lanthanum offering multiple benefits to manufacturing such as energy reduction and improved throughput. Experimental materials science and economic implications of Al-rare earth element alloys will be discussed. We show that Al–La/Ce alloys have elevated mechanical strength compared to more traditional aluminum alloys, in some formulations can be used without heat treatment, and possess a highly castable eutectic microstructure. This report presents the use of cerium and lanthanum in aluminum alloys as an example of how supply chain focused approaches to technological development can benefit stakeholders at every step in production.

Graphical Abstract



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Keywords Aluminum · Alloys · Rare earth elements · Critical materials · Sustainability · Cerium

Introduction

The production of rare earth elements (REEs) is fundamental to the growth of many modern technologies. Electric vehicles, wind turbines, and data centers are just some examples of high growth industrial sectors dependent on REE based high performance magnets [1]. Unfortunately, there is a persistent imbalance in REE mining. The main economic driving force for ore body exploitation (neodymium (Nd), praseodymium (Pr), dysprosium (Dy), and samarium (Sm)) is less than 25% of the overall REE content [2]. These desirable REEs are commonly mined from the same deposits as the byproduct REEs lanthanum (La) and cerium (Ce), which comprise greater than 70% of total production volume. Because REE mining requires sequential separation, production of La/Ce oxides cannot be avoided; and as such, what portion of La/Ce that is sold goes into low-value applications [3] with the rest returned to the mine as spoils at additional cost. One approach to address demand imbalance is to develop high-volume and high-value applications that increase demand for La/Ce. Growth in demand and value would shift the point of profit for rare earth mines toward the lighter elements, potentially lowering the price and/or supply volatility of Nd and other critical REEs [3].

The use of La/Ce as primary alloying additions in Al production is a possible application well-suited to promoting La and Ce demand. Commercially available Al–La/Ce alloys show they offer good manufacturability, high temperature mechanical stability, and in some formulations can be used without heat treatment [4–6]. Considering that Al alloy production exceeds 50 megatons per year (compared to 240 kt/y total global REE production [2]), even a small amount of La/Ce adoption in alloy manufacturing could drive demand to increase La/Ce prices. Concurrently, Al alloys are among the most energy-intensive materials in widespread structural applications, and their manufacture results in large emissions and power generation burdens. Heat treating of parts is the most time intensive and second most energy intensive step in foundry manufacturing, behind metal melting. Therefore, using La/Ce in Al alloys to reduce or eliminate the time and energy use associated with heat treatment will improve foundry economics, lower physical infrastructure needs, and shorten production timelines as demand for aluminum increases [7, 8]. Al–La/Ce alloys have the potential to simultaneously benefit the aluminum alloy and the REE mining industries. These benefits will be discussed individually before outlining the larger economic impacts and future outlooks.

Material and Methods

Conventional metal casting practices were employed to produce Al–Ce alloys. Permanent mold test bars were cast using the ASTM B108 standard from commercial commodity grade Al, Ce, and Mg all with purities above 99% in a 27 kg resistively heated furnace held at 760 °C. For the alloy, Al metal was melted prior to addition of Ce which was fully incorporated before Mg addition. A final nominal composition of Al–8 wt% Ce–10 wt% Mg was attained before the melt was cleaned with a halide-based flux. Note the alloy used in experimental assessment does not contain La, but recent work by the authors has detailed the viability of using Ce and La in combination or individually to achieve desired compositions and properties [9]. Therefore, for the remainder of this report the term Al–La/Ce will continue to address the broader family of alloys and Al–8Ce–10Mg will be used in reference to this alloy. Comparison alloys were either cast from master alloys using the same method or obtained from commercial sources in the case of wrought material.

Hot isostatic pressing (HIP) of Al–8Ce–10Mg alloy was performed at a temperature of 520 °C for 2 h and isostatic pressure of 100 MPa. Pressure was ramped along with temperature then retained during a slow cooling before being released.

Room temperature mechanical properties of as-cast and HIP Al–8Ce–10Mg were measured using a United model STM-100KN tensile testing machine with a constant cross-head speed of 6.5×10^{-3} per minute following the ASTM E8, Rev 88 standard for tensile testing of metallic materials. Elongation was measured to failure with United DATUM software and was confirmed by inspection following the test. Tests at elevated temperature were performed following the ASMT E21 standard.

SEM imaging was performed on a Hitachi S-4700 in backscatter mode with an accelerating voltage of 10 kV.

Gross value was estimated based on the product of 2019–2020 FOB China oxide prices and total recovered production for individual REE.

Benefit I: Coproduction of Metals

The demand for REE minerals has been steadily increasing, with global production rising from 80 kt of rare earth oxides (REO) in 2000 to over 240 kt in 2020 [10]. REE production growth is principally driven by demand for permanent magnets, which are central to the design of high efficiency compact motor/generators such as those found in hybrid/electric vehicles, direct drive wind turbines, and high

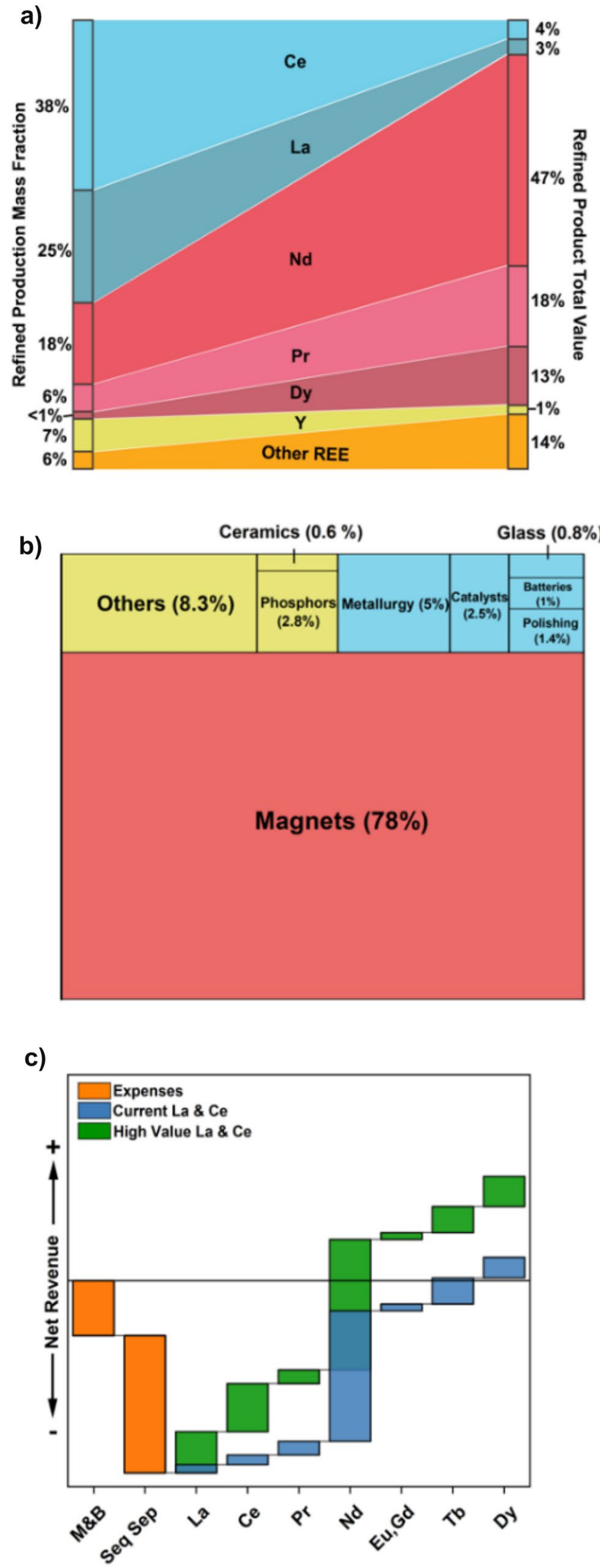

Fig. 1 **a** Allocation of REE production by weight compared to gross ▸ values [10–15], **b** Major REE markets by share of gross value, sectors are color coded to match largest contributing REEs form A [10–15]. **c** Illustrative plot of possible revenue shift after accounting for costs of mining and beneficiation (M&B) and sequential separations (seq sep) with factor of 5 increase in La/Ce value [10–15]

capacity HDDs [11]. REEs are naturally co-located within the same ore bodies as a single mineral, e.g. bastnaesite (REE-fluorocarbonate) or monazite (REE-phosphate). On average, La/Ce account for > 70% by mass of ore body content [12]. However, they are only in demand for relatively low-value industrial use in functional additives for iron production, catalysis, and polishing applications. As a result, they account for < 10% of REEs total economic value (Fig. 1a, b) [13–15]. Thus, La/Ce can be categorized as byproducts, goods with low net realizable value compared to main products. By comparison, the REEs used in magnet applications (Nd, Pr, Dy, Sm) often constitute less than 25% of ore body concentration but account for 78% of REE market value (Fig. 1a, b [13–15]). Reviews with more information on the specific processes and challenges of REE mining are available from Xie et al. [16] and REE recycling from Ginosar et al. [17].

The imbalance between composition and value places the economic burden of REE extraction on the low volume elements with high value applications since mine profit is completely tied to their efficient refinement and sale. Further, market reports and supply-chain analyses only account for the refined product reaching the market and do not include the substantial fraction of La/Ce remaining in the tailings that goes unprocessed due to low demand [18]. Therefore, Fig. 1a, b represents only published reports of REE production, and the portion of REEs composed of La/Ce and the resulting economic burden is likely higher.

Comparing La/Ce to other mineral byproducts and coproducts is instructive to contextualize the scale of the problem. Coproducts typically account for small percentage of a mine's output, are recovered in the latter stages of a production process, and would not be economically viable as an individual product. A classic coproduct example is indium (In), which is almost exclusively produced during the processing of zinc (Zn) and copper (Cu) ores. In 2013, 13,500 kt of Zn was produced globally with only 0.63 kt of In produced [19]. Zn mine profitability is not dependent on the small quantity of difficult-to-extract elements, but instead benefits from indium having ~ 100× the value of Zn per kilogram. Despite the high value, its low volume means In only accounts for about 0.5% of the mineral value produced in the mines. In REE mines, by contrast, this situation is reversed. They generate larger quantities of La/Ce oxide ($1.50/kg in 2020) compounds during initial steps of the concentration and separation processes, but their profitability is dependent on oxides of Pr ($60/kg), Nd ($47/kg), and Dy ($258/kg) that are separated later because demand for La/Ce is so low.

Supply imbalance is intrinsic to industrial REE production processes and cannot be avoided. Due to their chemical similarity, industrial REE separation is sequential, requiring the isolation of La/Ce first before separating heavier, more valuable REEs [20]. In comparison, the value imbalance is extrinsic to REE mining and solutions can be found. Creation of new high-volume and high-value demand for La/Ce in new industrial sectors could be a major step towards a resolution. Diversification of demand streams and new customer bases could also reduce volatility in REE mining, separation, and beneficiation because revenue is spread out instead of principally concentrated in a single application. As an illustration, Fig. 1c depicts a possible scenario where La/Ce $/Kg price increases by a factor of five above present value. The increase in value shifts the point at which positive net revenues are generated earlier in the separations process, reducing the economic reliance on the heavier REEs and cushioning production against price fluctuations.

Current demand for La/Ce is stable but dispersed across several low value applications where demand is elastic and readily available substitute materials limit potential price increase. Using large amounts of La/Ce in Al alloys could be a key step toward promoting the inelastic demand for La/Ce necessary to rebalance REE production and stabilize supply chains. However, to drive demand new technologies require a value proposition to incentivize adoption. The new Al–La/Ce alloys provide improvements to cost, processing, and performance compared to standard Al alloys [21–23]. This performance advantage allows them to compete against much more expensive alloys making them less sensitive to moderate price increases and expanding their possible application window.

Benefit II: Energy Reduction of Aluminum Alloys

The standard production timeline for an Al alloy part within a foundry (Fig. 2) includes a two-step heat treatment in which parts are held at elevated temperatures for 10 to 30 h in low thermal efficiency furnaces (solutionizing) before being quenched in water and heated a second time in a separate furnace (aging) [24]. During solutionizing (450–550 °C), solute elements are dissolved into Al and then retained in a supersaturated solid solution through quenching. The aging step (100–200 °C) then forms finely dispersed precipitates from the solutionized elements via age hardening. Tailoring the particle formation during aging allows for mechanical strength and ductility to be optimized for a given application [25].

In addition to the high energy demand in aluminum heat treatment, rapid cooling during water quenching can cause severe, often destructive distortion of parts due to thermal expansion gradients [25]. This distortion can prevent the use of certain cast alloys in many applications and higher casting rejection rates, causing waste or limiting design flexibility. In some cases, post heat treatment deformation may force manufacturers to add machining steps or forgo entirely the less expensive casting production methods in lieu of costly and time-consuming subtractive methods [22].

Al-La/Ce alloys are a new family of alloys compatible with numerous manufacturing methods including: sand and permanent mold casting; die-casting; extrusion; and additive manufacturing with equally broad possible application spaces [23, 26, 27]. Al-La/Ce alloys can include alloying additions like Si and Cu while retaining manufacturing flexibility [23, 26–28]. In contrast to most commercial aluminum

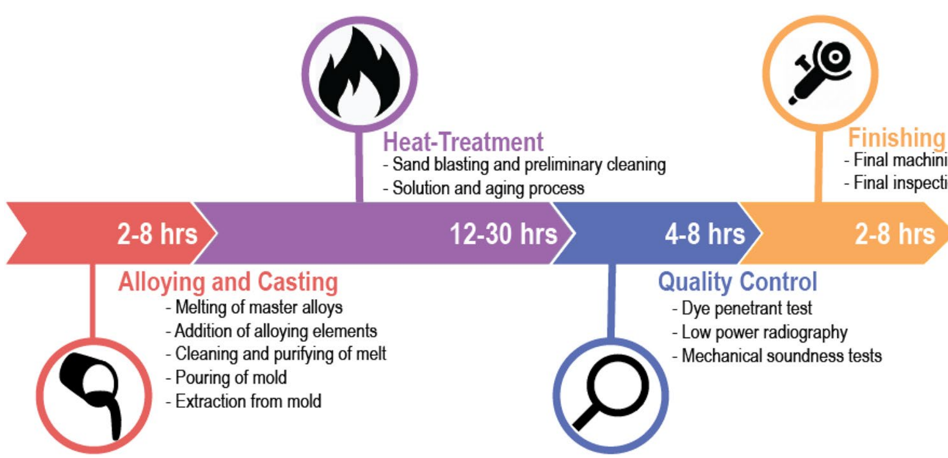

Fig. 2 Timeline of production for heat treated aluminium alloys within a foundry

alloy families like Al–Si and Al–Cu, Al–La/Ce are not reliant on solid-state precipitation to develop strength. Instead, Al–La/Ce alloy mechanical properties are principally tailored during casting through composition and process modifications (*e.g.* adjusting the amount or type of alloying element or increasing cooling rates) [4, 5]. Their solidification structure consists of a well dispersed, insoluble, thermally stable intermetallic, which for binary Al–La/Ce alloys is $Al_{11}(La,Ce)_3$, dispersed in an aluminum matrix. The two-phase cast microstructure imparts sufficient increase to room temperature mechanical properties and resists coarsening at elevated temperature, leading to its potential use in a broad range of applications [4, 5, 29]. By contrast, the intermetallics in common commercial Al alloys begin coarsening after high temperature exposure resulting in a rapid loss of strength which limits their utility for high temperature use. Further, solidification derived microstructural tailoring frequently enables Al–La/Ce alloys to be used without the need for heat treatment reducing the associated energy use and part distortions.

One promising example of this alloy class is found within the ternary Al–La/Ce-Mg system [4, 5]. The eutectic nature of Al–La/Ce alloys and lack of interaction between La/Ce and Mg during solidification enables substantial Mg additions (up to 10 wt%) while maintaining good castability. This can be contrasted to alloy 520 (Al–10 wt% Mg) which has very poor castability, causing difficult manufacturing and high rejection rates [30]. The principal features contributing to the elevated strength of Al–La/Ce–Mg alloys include high content of dissolved Mg in the Al matrix and a finely structured eutectic microstructure that forms during casting. These factors make cast Al–8 wt% Ce–10 wt% Mg (all further compositions reported in wt%) appropriate for use without heat treatment.

Al–8Ce–10Mg and comparison alloys were cast and mechanically tested in tension (Fig. 3). The selected comparison alloys are common in several elevated temperature applications for Al: numerous engine blocks are produced from A356 (cast Al–Si–Mg), pistons can be manufactured from 4032 (wrought Al–Si), and some turbine applications make use of 2618 (wrought Al–Cu) [29–31]. In the as-cast state and at room temperature Al–8Ce–10Mg possess higher yield strength than heat treated A356, though lower than wrought alloys (e.g. forged, rolled, extruded) 4032-T6 and 2618-T6 [32, 33]. This changes dramatically with temperature where, for example, at 200 °C Al–8Ce–10Mg possesses more than double the yield strength of competing materials. Further, the precipitate strengthening mechanism in traditional alloys is unstable at elevated temperature causing these alloys to permanently lose a portion of their strength when returned to room temperature following such thermal excursions [34]. This is not the case for Al–La/Ce alloys

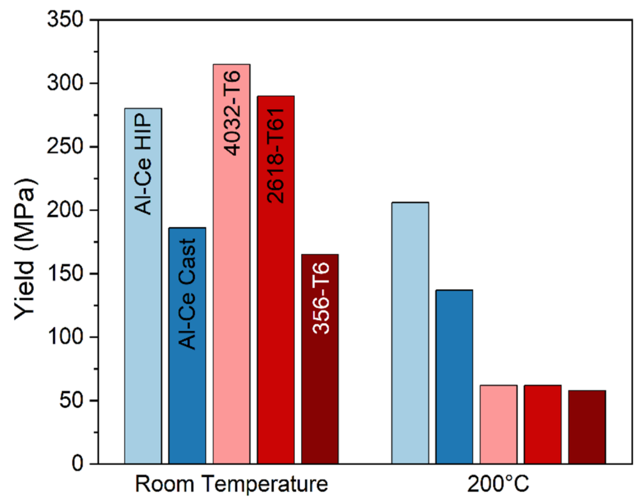

Fig. 3 Comparison of as-cast and HIP Al–8Ce–10Mg alloys against more traditional cast and wrought aluminum alloys at room and elevated temperature (200 °C) [27]

which, after elevated temperature exposure typically retain their full strength [29].

The reduction or elimination of heat treatment afforded by the use of Al–La/Ce alloy may also be attractive to high-performance Al alloy sectors. Hot isostatic pressing (HIP) is another less common thermal treatment where a pressure vessel is loaded with castings, pressurized to 50–300 MPa (~500–3000 atmospheres), and heated to above 450 °C [35]. This process relies on combined high temperature and pressure to soften alloys and then heal casting related defects by closing internal voids and redistributing soluble elements (homogenizing) rapidly [35, 36]. HIP processing is not a stand-alone thermal treatment. Alloys which undergo HIP treatments also require secondary processing steps (i.e. solution and aging heat treatments). The high-value applications requiring HIP often possess rigorous design and geometric tolerances, amplifying the effect of distortions that may occur during the quench step of the solution-age heat treatment [24, 35, 36]. Therefore, reduction or elimination of the post-HIP thermal treatment would not only lower the energy expenditure but may offer a means to retain the full benefit of HIP processing [37].

During experimental trials on Al–Ce–Mg alloys, a HIP treatment was applied without additional heat treatment and resulted in improved microstructural homogenization, mechanical strength, and elongation to fracture. Microstructural comparison between as-cast and the HIP alloy (Fig. 4) shows that the application of a HIP treatment is effective at homogenizing soluble elements quickly, yet the fine reinforcing phases are preserved (Fig. 4a, b). Despite the high temperature of the process, the $Al_{11}Ce_3$ intermetallic did not change morphology or coarsen (Fig. 4a–c), and grain growth, thermally activated coarsening of the matrix phase,

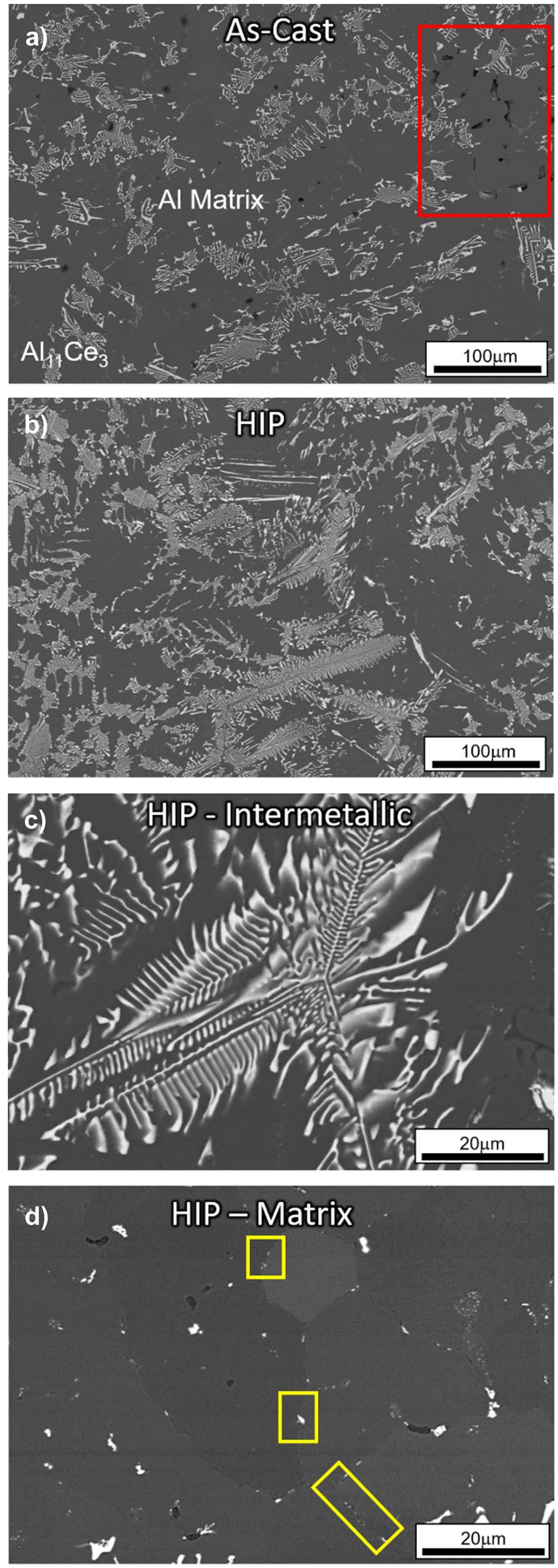

Fig. 4 SEM backscattered electron micrographs showing the microstructures of as-cast and HIP processed samples of Al–8Ce–10 Mg alloy being composed principally of Al–La/Ce intermetallics (bright white areas) inside an Al-rich matrix (gray areas), **a** as-cast Al–8Ce–10 Mg with, red box outlines area of mixed voids and Al–Mg β phase which forms in a divorced nature as the final step of solidification, **b** HIP condition with a visually equivalent microstructure to the as-cast condition—note the absence of voids, **c** HIP condition at higher magnification revealing a stable intermetallic structure that does not dissolve or coarsen, and **d** the Al matrix with discrete intermetallic $Al_{11}Ce_3$ particles decorating the grain boundaries (yellow boxes) (Color figure online)

was not observed after HIP. Small Al–La/Ce intermetallic particles at grain boundaries (Fig. 4d) prevented grain growth through grain boundary pinning, the process by which grain growth is halted or slowed by impingement of grain boundaries on another phase. The observed increase in mechanical properties of the HIP alloy over the cast (Fig. 3) likely resulted from solutionizing of Mg that had resided in the now dissolved β-phase ($Al_{140}Mg_{89}$) that formed during solidification. Additionally, a more homogenous microstructure and elimination of voids results in the ductility increasing from ~ 1% in the as-cast state to 10–12% in the HIP condition.

Potential Demand Growth and Market Impact

Decreased manufacturing time, energy savings, and improved high temperature performance could incentivize Al–La/Ce alloy adoption. In many cases, these alloys can be adopted without the need to change legacy manufacturing methods. For example, the vast majority of cast aluminum production is dedicated to Al–Si alloys, and Al–La/Ce alloys can be directly exchanged for Al–Si production pathways without retooling. The broad compatibility of Al–La/Ce alloys with casting, wrought processing, and additive manufacturing techniques further expands adoption pathways.

Significant energy savings from Al–La/Ce alloy production will accrue with large adoption volumes. For example, engine components such as diesel cylinder heads and engine blocks that are commonly produced from heat treated Al–Si alloys could create high-volume demand for Al–La/Ce alloys which possess superior performance at high operational temperatures. The energy expenditure required to heat treat sand cast Al engine cylinder heads can reach 2150 kWh per metric ton of finished products (Fig. 5) [7]. Model demand growth of Al–La/Ce alloys for automotive drivetrain components predicts a possible market size of 280 kt for Al–8La/Ce alloy by the year 2055 [38]. Thus, using these adoption scenarios in combination with data on foundry energy saving from heat treatment reduction, it is predicted that broad use of Al–La/Ce alloys could by 2055 save between 425,000 and 675,000 MWh annually. This is the approximate energy consumption of 60,000 US homes [7, 38]. At the same time it would provide a new demand stream for La/Ce of 22.4

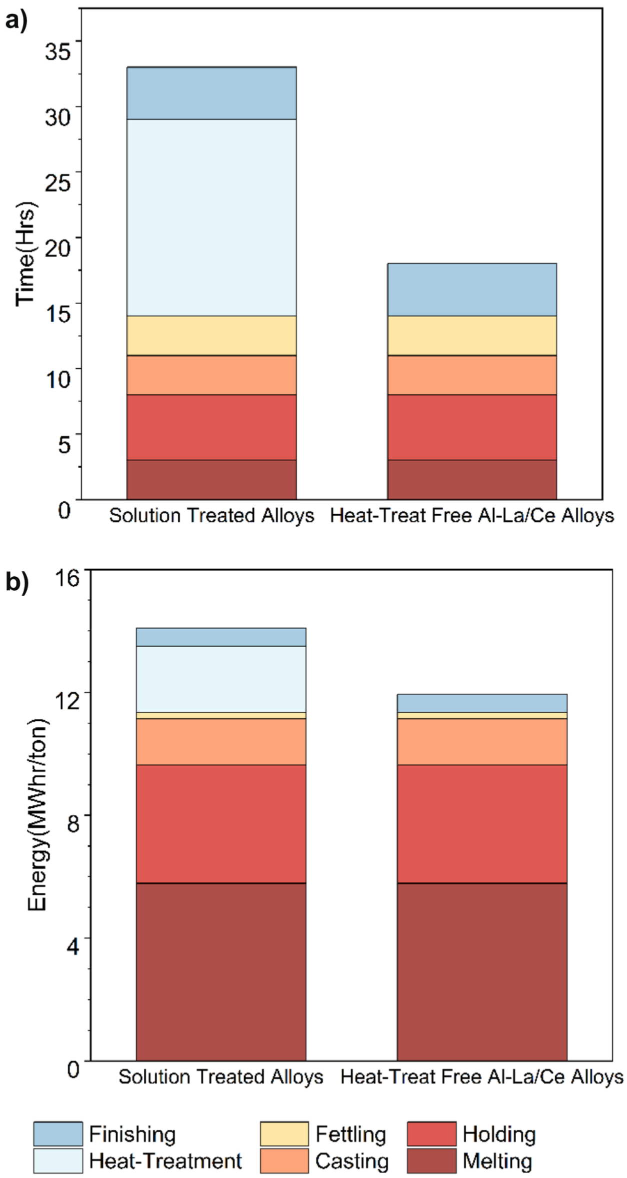

Fig. 5 Plots of potential for decreased **a** manufacturing time and **b** energy expenditure of Al–La/Ce heat-treat free alloys compared to solution treated alloys. Removal of heat treatment leads to an energy decrease of approximately 16% with a corresponding decrease in CO_2 emissions. Fettling is the process cleaning parts of extra material after removal from the sand or permanent mold

kt/y in only a single application. Comparatively, the global production of REEs in 2020 was 240 kt (70% being La/Ce) [2]. The reduction or elimination of heat treatments also decreases production timelines and reduces manufacturing footprint.

Growing demand and price for byproduct REEs would increase mine viability by motivating in house sequential separations. Demand growth of the least valuable REEs helps to stabilize existing production, develop additional ore sources, and incentivize non-traditional production, thereby diversifying supply [39]. Al–La/Ce alloys have the potential to create and sustain a necessary level of demand for the prices of low value REEs to increase, shifting the point of positive net revenue forward in the sequential separations process.

A significant concern for Al–La/Ce alloy early adopters is a stable supply chain. Current technology requires the initial separation of La/Ce from ore deposits to enable further separation of the valuable elements. The lack of market demand has reinforced the mine operators' habit of discarding or stockpiling La/Ce. These extant stockpiles will enable producers to adapt to near term La/Ce demand growth, and ramp production to meet future demands. Increased prices from higher demand will push the price of Al–La/Ce upward toward alloy families like Al–Mg improving global REE supply chain stability. The lower manufacturing costs and improved alloy properties make Al–La/Ce alloys competitive with higher cost Al alloys and potentially against some Ti alloys.

Outlook

Global supply chains are an indispensable part of the world economy, and disruptions are frequent throughout history. Further, because material needs are dynamic with discovery of new applications effecting demand, supply chains that were robust can quickly become volatile if not correctly adapted. In the early to mid-twentieth century, REEs were not used in particularly critical applications (uses included polishing compounds, lighter flint, and fireworks) and supply disruption was not a risk. This changed with the development of rare earth permanent magnets and other energy conversion technologies in the late twentieth century, making the REE supply chain essential for many consumer industries. REE supply chain expansion and diversification could not grow at the same pace, and the impact of supply shortage was realized in 2011 when a massive REE supply reduction increased the price of commodity grade REE by over 2000%, sending economic shockwaves through many industries that relied on REE supply. The 2011 REE price spike was caused by factors other than crustal abundance, showing how supply chain risk can arise unexpectedly.

The COVID-19 pandemic has revealed the potential of choke points in complex distribution networks to cause cascading failures across entire economies. Diversification of supply sources is a viable method to mitigate this risk. For instance, REE supply is heavily concentrated in a single region despite globally distributed resources. In many cases miners are reluctant to exploit new ore bodies because of their low concentration of the valuable elements needed for magnets. Emerging demand for Al-La/Ce alloys has the potential to make new REE mining economically viable for those deposits less rich in elements used for magnet production. This pathway towards diversification of REE supply chains would build in natural resilience to supply shocks (temporary but extreme disruptions) as they become more likely due to external factors such as climate change and environmental disasters.

Availability of new, fully qualified material systems to be injected into compromised supply chains can also support resilience in the case of disruptions. For example, the price of metallurgical silicon spiked by 400% between June and November of 2021 [40]. Severe flooding in China, which produces ~ 70% of the world's silicon, restricted coal extraction and increased energy costs to the point where Si production profitability was impacted, and output was reduced [40, 41]. Si is a key ingredient in many of the most important Al alloys and was an unexpected source of supply uncertainty to casting houses. In this case, were casting houses to have access to fully qualified material systems that could replace Al–Si in some applications like Al–La/Ce alloys a portion of the supply risk could be mitigated.

To reach large scale adoption levels Al–La/Ce alloys have barriers to overcome before adoption can expand rapidly. For example, modern alloys are often tailored to an application by leveraging existing knowledge bases about alloying element interactions. However, the recency of Al–La/Ce alloy development means the interaction of alloying additional elements with Al–La/Ce alloys is not yet well understood, and active research is needed to develop this knowledge base. Another possible barrier is feedstock transportation and cost. Ce and La must be transported as flammable material and are expensive compared to elements like Si that find wide industrial application. Though, new techniques for manufacturing by directly reducing the oxides or carbonates of Ce and La in molten aluminum are being investigated to overcome these cost and transportation challenges.

Though much work remains in exploring the family of alloys that make use of La/Ce as primary alloying additions, there have been several exciting discoveries regarding La/Ce additions to existing Al alloys. Ce additions can improve the morphology of Fe containing intermetallics that are known to reduce alloy ductility. Also, Ce additions to Al–Si alloys scavenge Cu impurities thereby increasing impurity tolerance and corrosion resistance in Al–Si alloys [42]. This result alone could drastically improve recyclability of an entire family of Al alloys.

Conclusion

Al–La/Ce alloys are just one example of how diversification of material technologies can improve supply chain dynamics for both up and downstream stakeholders. They reveal how a supply chain focused approach to technology development can increase resilience, sustainability, and economic viability. Specific to this system, Al–La/Ce alloys can exploit untapped value in the REE supply chain and in aluminum alloy markets. Demand growth potential for these alloys is high and can be directly inserted into current manufacturing pathways while reducing energy costs. There are still impediments to overcome in their pathway toward adoption, but when realized the eliminated production steps can benefit the spectrum of high-performance Al applications without sacrificing mechanical properties, weight savings, or design flexibility. Additionally, the broad applicability will induce higher demand for Al–La/Ce alloys and La/Ce as additions to current commercial Al alloys, in turn, increasing demand for abundant La/Ce adding stability to this critical material supply chain.

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