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Alloy design via additive manufacturing: Advantages, challenges, applications and perspectives

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Additive manufacturing (AM) has rapidly changed both large- and small-scale production environments across many industries. By re-envisioning parts from the ground up, not limited to the challenges presented by traditional manufacturing techniques, researchers and engineers have developed new design strategies to solve large-scale materials and design problems worldwide. This is particularly true in the world of alloy design, where new metallic materials have historically been developed through tedious processes and procedures based primarily on casting methodologies. With the onset of directed energy deposition (DED) and powder bed fusion (PBF)-based AM, new alloys can be innovated and evaluated rapidly at a lower cost and considerably shorter lead time than has ever been achieved. This article details the advantages, challenges, applications, and perspectives of alloy design using primarily laser-based AM. It is envisioned that researchers in industry and academia can utilize this work to design new alloys leveraging metallic AM processes for various current and future applications.

Keywords: Alloy design; Additive manufacturing; Directed energy deposition (DED); Powder bed fusion (PBF); 3D Printing

Introduction

Metal additive manufacturing (AM) has seen extensive interest in the past decade from manufacturers in the biomedical, aerospace, energy, and nuclear sectors, among many others [1–6]. The ability to create components with unique structural and compositional characteristics not possible using traditional manufacturing methods has led to an expansion of interest among engineers and researchers alike for both lab-scale material and structural innovation and large-scale part production environments. In recent years, we have observed AM's impact on improving the existing performance and supply-chain characteristics in many different areas. More importantly, however, manufacturers and researchers are beginning to show us how the technology can transform the future by adjusting entire materials-design-manufacturing processes compared to traditionally accepted ones. More specifically, the AM technology platform is changing how different heritage companies across numerous industries can design and manufacture structures to increase complexity, customization, and consolidation to enhance efficiency and functionality [7]. Further, a recent Gartner report [8] estimates that three of the most cited reasons for adopting AM technology are – (i) prototyping, (ii) product development, and (iii) innovation – indicating that companies are heavily invested in the future development of products utilizing AM. Several recent reviews have cited how a shift in AM intellectual property (IP) and “fragmented” individual research in specific areas will become more centralized within a decade, enabling

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an improved future in the materials-design-manufacturing space [9,10]. Additionally, AM has shown the world that a rapid response to a global pandemic such as COVID-19 in the form of face shields, respirators, and other necessary items is possible. Unlike in the past, material development and manufacturing are no longer a bottleneck to the design-manufacturing process in dire situations [11,12].

In metallic materials specifically, AM has shifted how we can envision new alloys for special applications. Based on AM's unique processing capabilities, new developments in this area are among the most exciting aspects of the next wave of innovation in the field. These efforts build on developing processing-structure–property relationships among pre-existing alloys transitioned to AM, which has been the primary focus of manufacturers with several review articles in the literature [4,13–15]. Among desirable characteristic features of laser AM (LAM), high cooling rates and the ability to melt and shape components from elemental powders traditionally requires high-temperature arc-melting facilities as well as subsequent processing steps. Because of this, achieving alloy development capabilities using LAM offers a significant supply-chain advantage for corrosion-resistant, refractory, and high-temperature materials necessary in industries such as biomedical [16,17], aerospace [18,19], and nuclear/energy [20,21].

Fig. 1a highlights a typical alloy design workflow where the initial conceptualization and need for the alloy are brought forward, the actual chemistries are then decided, and the two primary metal AM methods, directed energy deposition (DED) and powder bed fusion (PBF), can be utilized to manufacture components from those materials. A discussion of these processes' mechanics will be highlighted in this review, and further reading can be found in references [22–24]. The exciting opportunities offered by LAM are just starting to be realized by engineers and scientists in the alloy design field, motivating a review of what has been accomplished in recent years, the key advantages of leveraging AM for alloy design, current challenges, and what is envisioned to drive the field forward in the future. To this end, this article combines necessary insights both from academia and industry, beginning with the motivation for performing alloy design using metal AM, namely, the ability to work with high-cooling rates, reduce tooling capital involved with a traditional alloy design, and the ability to innovate for specific applications; discussion on what key advantages and challenges manufacturers and researchers have faced; namely, previously developed alloys are not always best for metal AM, overcoming a production-centric barrier on new material innovations and designs; and finally discuss past and current works on development of new aluminum and titanium alloys, steel alloys, high-entropy alloys, and magnetic alloys that exhibit enhanced properties and characteristics over existing materials. We shall conclude with a critical look at the future trends and challenges envisioned, involving modeling tools and developing functional gradient structures for critical applications. With the rapid rise in the literature related to metal AM, manufacturers need to have a path to adoption of this technology that combines insights and perspectives from both academia and industrial professionals. It is envisioned that this review will inspire the next generation of materials engineers and scientists seeking tools and understanding of how to leverage AM to develop the world's next alloys to meet tomorrow's needs in numerous applications.

Current need in alloy design

Pure metals are rarely used in any application as their properties are not suitable or tailored to a product's specific needs. But a small addition of a 2nd or 3rd element to a pure metal can cause a significant change in the properties of the alloy. A simple example is different steels. Just a small addition of carbon to iron makes steel, and depending on the amount of carbon addition — the properties of steel can be very different. Then, further adding other alloying elements can make steels either stainless (with chromium addition) or tool steels with high hot hardness (with different carbide formers such as vanadium, molybdenum, etc.) precipitation hardened steels for various applications. Similarly, the addition of alloying elements can make steel either magnetic or non-magnetic at room temperature. Designing new alloys happens based on the application needs, and it requires extensive experimental capabilities, from high-temperature furnaces to melt different elements to post-processing equipment for shaping. Becaise of this, for several decades significant efforts have been devoted to surface modifications of an alloy rather than redesigning new alloy chemistries because surface modifications are easy to do where bulk properties are not compromised. A simple example is biomedical devices. The entire metallic biomedical device industry primarily uses three different alloys – stainless steel 316L, titanium and its alloy Ti6Al4V, and CoCr alloy. Although a few other alloys have found exotic applications, such as nitinol and magnesium alloys, their volume consumptions are low. For load-bearing implant applications, Ti6Al4V is very popular. However, the osseointegration of Ti6Al4V is limited. Various coating systems have been designed to enhance the osseointegration (or biocompatibility) of the Ti6Al4V alloy, including calcium phosphate coating, tantalum metal coating, or porous titanium coating. However, minimal effort has been devoted to redesigning a new Ti-alloy with better biocompatibility. It is also important to note that Ti6Al4V was never designed for biomedical devices, but for aerospace applications. Excellent fatigue and corrosion resistance of Ti6Al4V alloy was the main reason for its original use in biomedical devices. The challenge is not just designing a new alloy because finding a vendor to manufacture those compositions on an industrial scale is also essential. And the solution is – keep using the legacy alloys and only make surface modifications to meet various anatomical and biological needs. However, alloy design via AM can make a significant difference because new chemistries can be invented using this approach and manufacturing functional parts with different shapes becomes possible. It is envisioned that the AM platform will be used to innovate new alloy chemistries for biomedical, aerospace, nuclear, and other performance-critical applications where there is a significant need for new materials. It is also important to note that although alloy design can be done via AM technologies, certification of those alloys for specific applications will still require extensive characterization.

Additive manufacturing-based alloy design

High-value, complex components such as biomedical implants, aerospace engine components, nuclear reactor parts, among

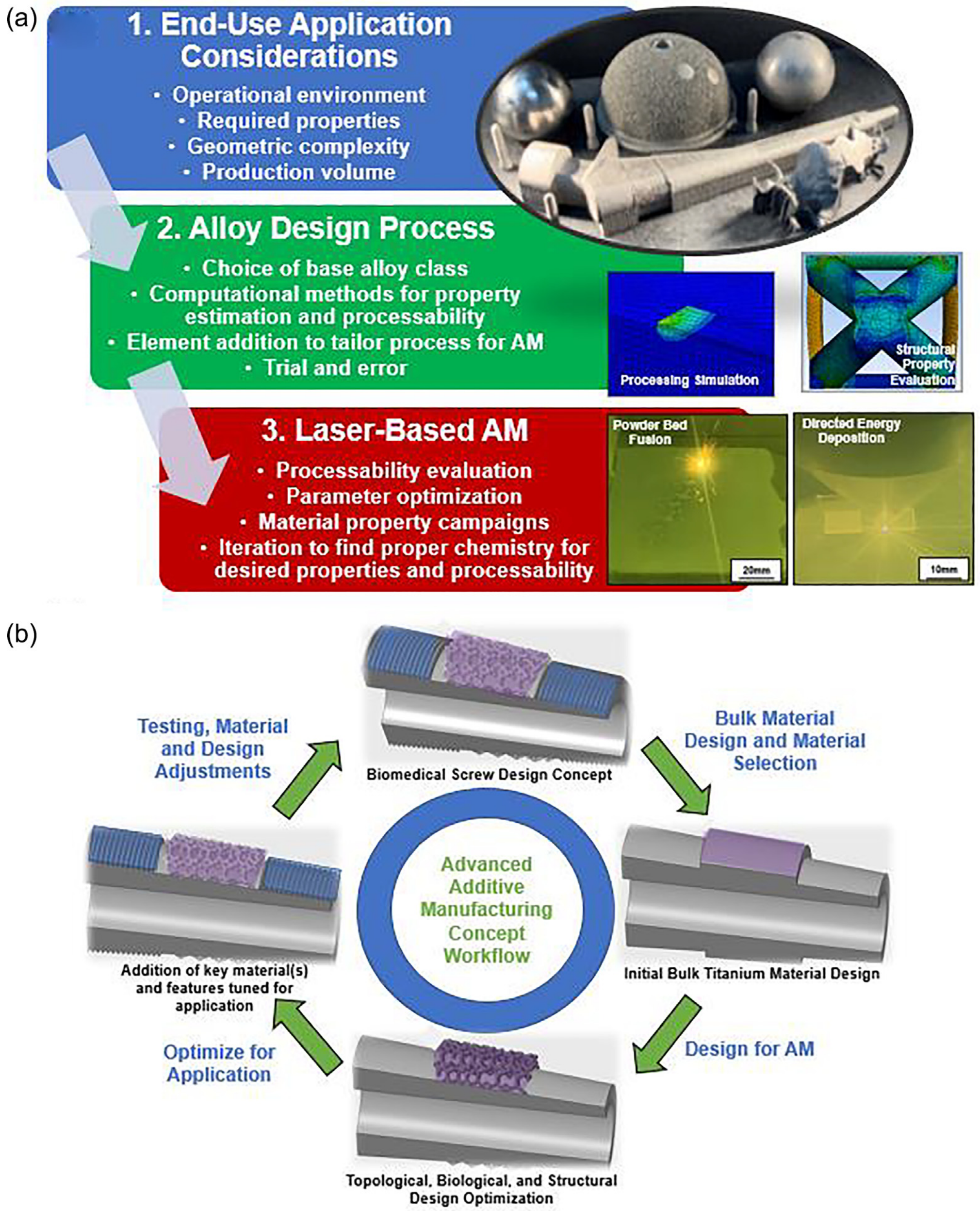

FIGURE 1

Example of current alloy design approach using laser-based additive manufacturing. (a) General workflow for incorporation of alloy design in a manufacturing process. (b) An example implementation of stated workflow towards a standard biomedical screw application.

many others (see Fig. 1), require specialized manufacturing techniques and properties to meet the application needs. Fig. 1 displays several examples of biomedical parts produced via LAM that would be otherwise challenging or too expensive to machine or cast using traditional processing methods. An example of this is shown in Fig. 1b, whereby a bulk component will be envisioned from new or existing materials, specific design optimization for the geometry, and site-specific features of new alloys tailored for the end-user. Standard design feedback is then applied where manufacturing challenges, or testing results of a component can lead to changes easily applied using additive-based methods, whether chemical, geometric, or functional. These adjustments can be defined through testing campaigns or process monitoring aided with machine learning-based programs that can aid processing optimization and reliability based on measurable properties and process metrics for understanding inputs to the additive process. These types of applications motivate heavy investment towards developing additive-based production methods to reduce overall costs and supply chain complexity across many industries. Some key example areas are shown in Table 1. Because processing-property relationships are integral to any design-production strategy, new alloy chemistries and designs are becoming apparent in the AM community owing to the unique processing characteristics of LAM, namely, high cooling rates and complex temperature profiles that might generate variable microstructures along with thermal residual stresses

TABLE 1

Application areas for alloy design approach via metal-based AM.

Material System	Industries	Design & Application Areas
Inconel (Ni–Cr)	Aerospace, Energy	Modification and/or enhancement of reinforcing phases for increasing temperature capability, oxidation resistance, fatigue, and fracture properties at high temperatures.
Titanium	Biomedical, Aerospace, Energy	Increasing biocompatibility, fatigue performance, and strength, corrosion and oxidation resistance
Aluminum	Aerospace, Energy	Increasing strength, fatigue performance, and processability via laser-based AM
High Entropy Alloys (HEAs)	Aerospace, Energy	Modification of strength/toughness, ferromagnetic properties, increasing oxidation resistance

within the parts during printing [24]. These characteristics can result in as-printed properties that are significantly different depending on the specific alloy family, trace element amount, and sometimes the processing parameters and orientation of testing relative to the build orientation of the AM process [25,26]. Further, the desire to modify the chemistries of existing alloys for processing's sake has naturally raised the question of what could be achieved by altering the material's chemistry for increased end-use performance. Most existing alloys used extensively in the industry have been developed for decades and have stood the test of time for reliability and suitability for specific applications. However, with the ability to rapidly create an extensive library of chemistries and properties for new alloys, LAM methods have opened the door to an unprecedented amount of innovation in the materials community.

Comparison of traditional alloy design process vs. additive-based alloy design process

Fig. 2 outlines a comparison of alloy design using traditional methodologies, such as casting, to additive-based approaches. Generally, the need for a new alloy can be envisioned from a specific and/or range of applications that would require a material system different from what is currently used, having improved properties compared to what exists in the market or a given supply chain. In many instances, the cost of the manufactured product is also a significant factor behind alloy design, such that the new alloy can perform the same task using low-cost and readily available starting materials. The new alloys may be designed via computational approaches, extensive experimental data, or first principles [27]. It is acknowledged that the development of a cast product is usually best accomplished using casting-based alloy design approaches, and the same for AM-designed products, owing to the significant difference in cooling rate, feedstock material, and scale, among other key metrics. As the motivation for more complex products for specific applications is growing, the use of additive-based approaches to alloy design is also becoming popular [22]. Thousands of parts have been produced and certified within the aerospace and biomedical industries. Despite these successes, lessons learned from casting-based alloy design regarding the choice of material chemistry, thermal–mechanical aspects, production volume, etc., have greatly influenced the current AM-based material development landscape.

An essential factor for alloy development is the extent to which researchers need to perform in-house alloy design and characterization, as most alloy procurement occurs from specialized vendors that can provide the specific chemistry desired in an ingot form or as rolled/forged products. As shown in Fig. 2, an organization provides the desired chemistries to the vendor to receive cast or rolled products back. Making new alloys is an extensive high-temperature operation. The new ones need to be processed in small batches, which poses a block in a production line for existing products. Such operation may also require unique hardware or environment to create a good ingot for the customer, such as vacuum-based processing or multiple/complex melt sequences. This may result in an eventual long lead time for special ingot from vendors and even further lead times for transitioning an alloy to an actual product. It is also important to note that such an alloy design approach requires many starting materials to produce the cast ingots. This is of particular concern for expensive starting chemistries like tantalum, nickel, or niobium, to name a few. In comparison, utilizing a powder additive-based approach relies on obtaining the raw elemental powders and having the AM equipment to create end-use parts with the same or varying chemistries. It is acknowledged that in the case of powder-based AM approaches, atomization is also typically a required step after the production of an ingot. However, with the increasing adoption of the technology and the many forms of atomization and powder morphologies capable of producing high-quality components, the costs will continue to drop for implementing an additive-based approach. Additionally, more challenge is involved in the storage of highly flammable metal powders than ingot or wire, which might be used in arc-melting based approaches. Among other characteristics, the additive-based approach ensures that all intellectual property (IP) regarding specific alloy chemistry and processing details are kept in-house, a substantial competitive advantage in high-value industries such as aerospace and biomedical. In this case, the only factor external to the company is the availability of the materials of interest, including rare-earth or other high-value alloying additions that may take some time to procure. Regardless, the vendor using the traditional approach will face the same issue but will likely require ample time to deliver the ingot product back to the company due to demands from other companies and the eventual long lead times incumbent upon foundries. From this point forward, in both cases, extensive characterization and testing of physical, mechanical, and thermal properties and other post-processing trials are incumbent upon the company to evaluate the suitability for the end-use application. The main goal is to down-select the several sets of chemistries sent out for production at this stage. Suppose none of the chemistries prove suitable for use; a design feedback loop must develop different chemistries. In that case, this poses a significant problem for the traditional approach as it calls for a complete re-run of the process from the beginning. Different chemistries could simply be processed in-house using the additive approach's

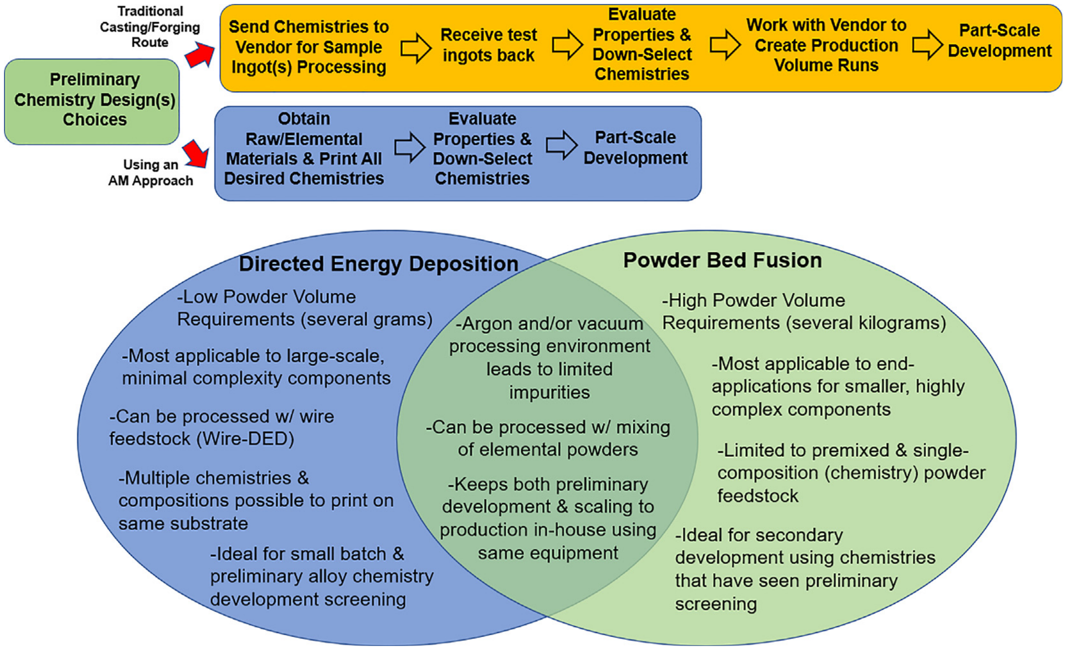

FIGURE 2

Comparison of alloy design approaches using traditional methods and the main metal-based AM methods – directed energy deposition (DED) and powder bed fusion (PBF).

existing setup, with limited time to reach the testing stage again. In either case, once the suitable chemistry is chosen, part-scale development is the next step, which is the most direct in the case of the additive approach as the alloy chemistries themselves were developed using the same machine from which parts will be produced. Comparatively, for the traditional approach, the vendor's existing setup may not suit the company's component needs in terms of complexity and volume. They may need to go through the same process with another vendor to meet production requirements as necessary for the end application. From this perspective, an additive-based alloy design approach saves significant cost and time relative to a traditional process used for casting-based products, offering a reduction in the supply chain and direct access to rapid design feedback loops as well as part-level production scaling.

Reducing lead times and cost using additive-based alloy design approaches is a significant motivating factor for breaking down the new materials' entry barrier. The high cost of researching new alloys, outsourcing chemistries to vendors to create new ingot for studies, and the time between each of the steps, each act as a significant deterrent to manufacturers on developing new materials, requiring only dire circumstances to motivate the investment of engineers and researchers to come up with the following best alloys. To add to the challenge, alloy design is a very high-risk endeavor as there is simply no guarantee that a new alloy will make it from the research stage to production within a reasonable or necessitated timeline, pushing engineers towards redesigning components with existing materials without the desirable properties or characteristics a new alloy would exhibit. Cutting down on the length of this feedback loop by leveraging LAM greatly alleviates cost and headaches down the road for engineers and researchers, motivating the exploration of new design spaces for alloys with different chemistries and characteristics than those currently available.

Table 2 summarizes the differences between AM-based and conventional alloy design approaches. Perhaps the most critical point to note is that the time has come to do alloy design for AM-based processes with high cooling rates than legacy alloys designed for conventional manufacturing approaches. As the AM approaches use legacy alloys such as Ti6Al4V, Inconel 718 without any compositional modifications, it is envisioned that the new alloys designed via AM can also be used in conventional manufacturing with minimum alterations of its chemistry. Finally, it is envisioned that innovation in alloy chemistry will solve long-standing challenges in various applications, such as designing alloys for fracture management devices without using Ni instead of 316L stainless steel (having 10% Ni) that is currently being used and can cause metal ion sensitivity to many patients. Similarly, designing Ti-based alloys that can be used beyond 450 °C for aerospace applications. Such innovations in designing new alloy chemistry via AM and other innovations such as topology optimization will redefine the future of manufacturing in the coming decades.

Towards achieving alloy design via metal additive manufacturing

The main metal–AM methods, namely DED and PBF, can be used for alloy design with several critical distinctions in the process itself as well as the available raw feedstock materials. Fig. 3 highlights different sub-categories of DED and PBF, with the most significant difference between the two main categories being the position of the feedstock concerning printing of the overall structure. More specifically, DED is a wire or powder-fed method that

TABLE 2

Key differences of metal alloy design using traditional vs. AM-based.

Aspect	Key Comparisons
Supply Chain	– The design feedback loop can be very agile using LAM due to most machine setups existing in-house and near the production line. – Scarce or rare-earth alloying elements that are highly expensive to procure in powder (or wire) form can limit exploration of highly exotic alloys using LAM compared to traditional methods that may not require powder (or wire) feedstocks.
Microstructure and properties	– Cooling rate differences can lead to different challenges and strategies for strengthening and controlling microstructure in traditionally as-cast vs. as-printed parts. – Higher cooling rates in LAM can lead to higher strength and lower elongation in many alloy systems left in the non-heat treated state.
Geometric Capability	– Scalability and desired geometry can impact the design of alloys and how practical an alloy design approach is towards a particular application. – Alloy design using LAM maintains all of the same advantages that single-material LAM possesses, i.e., complex internal geometries and the ability to process one-off parts for advanced applications, which lends itself well to advanced alloy design only being needed for several parts. – Traditional processing via investment casting of new alloys is well suited for high volume production; even alloys developed using LAM may be able to be transferred to traditional casting-based approaches.

utilizes an inert gas flow (typically argon, and in some cases nitrogen and vacuum in the case of e-beam) to direct powder feedstock (spherical powders typically in size range of 50–150 μm in diameter) or wire into the melt-pool that is at the focal point of a laser (or electron beam) above the build platform [28]. To create shapes using this method, the build-plate or the powder flow head can move in different directions, with the option of utilizing 5-axis or free-axis control of a hybrid machining center add-on, which adds a subtractive capability to the additive process [29].

Feature resolution for DED ranges from 200 to 500 μm depending on the specific processing parameters, energy input, spot size, and operational characteristics. One of the key advantages of using this AM method for alloy design is the relatively small mass of powder required to print a structure large enough for analysis and testing (see Figs. 3 and 4a), with further reading found in ref. [30,31]. Further, the ability to utilize multiple powder feeders to mix different chemistries in a single print allows for testing a wide array of chemistries with slight alterations to the trace amounts of alloying additions. For example, FormAlloy's Alloy Development Feeder (ADF) with a 16-alloy element hopper allows for extensive parameterization in deposited material composition [32]. More specifically, when processing custom compositions with DED, there are two key strategies: (i) on-the-fly alloying and (ii) premixed-powder alloying. On-the-fly alloying allows the user to enable the desired powder feeders with defined parameters to send material through the powder transport lines and into the deposition head. Several powders can be sent through the lines and experience “blending” as they are transported into the deposition head. Such an approach allows the user to load powder feeders once and control the powders' delivery into the deposition head and subsequently to the melt pool. However, the challenges of on-the-fly alloying might outweigh the advantages in certain applications. These challenges include consistency of powder flow, the complexity of powder compositions, the need for a large number of feeders to address all required elements, and perhaps the biggest hurdle is adjusting the feeder flow rates to achieve desired compositions as the final chemistries are subject to perturbations in the overall flow system, as well as the rheology of the constituent materials themselves [32]. Also, during on-the-fly alloying, all unused powders get mixed and may not be reused again. However, despite these challenges, Moorehead et al. (2020) demonstrated fine-tuning of a DED system for on-the-fly alloy approach and achieved within 5-10 at% a multi-component high-entropy alloy system [33]. On the other hand, premixed-powder alloying addresses several issues of on-the-fly alloying, but still requires many powder feeders depending on the alloy processing strategy utilized. Dippo et al. have shown that the as-deposited phase percentage using premixed alloying methods can closely resemble the desired/predicted composition with a multi-feeder system [32].

Most powder feeders come with two components:

1. The powder hopper that contains the feedstock material.
2. The powder delivery base that controls the amount of powder leaving the hoppers via a delivery mechanism (usually rotary) and control of the argon/nitrogen “carrier” gas flow through the hopper.

Using the premixed alloying approach with traditional powder feeders, users could have as few as one delivery base and as many powder hoppers as they would like. The advantages of having premixed powders in separate hoppers allows for a high degree of control and accuracy of the deposited samples' overall chemical composition and homogeneity compared to an on-the-fly strategy whose final chemistry is subject to many factors. The challenges with this approach are the additional powder hoppers' cost and the time required to manually change each hopper's material to print different alloy compositions. The demonstration piece in Fig. 4a shows that many different chemistries can be printed and post-processed on a single substrate and with comprehensive microstructural and properties characterization (e.g., porosity, grain structure, and mechanical properties) [32]. These characteristics lend DED nicely towards alloy design studies with a wide range of desired chemistries, where applicability to an end-use part (tailored for a powder-bed process) is not of immediate interest, i.e., first-generation studies. Further, wire-based DED, such as wire arc or plasma arc AM, can be utilized to create new alloys by inserting multiple alloy rods into the melt pool or inserting a rod of previously melted ingot [34,35]. This technique's main advantage is the availability of various commercial feedstock rods for alloying, but this also

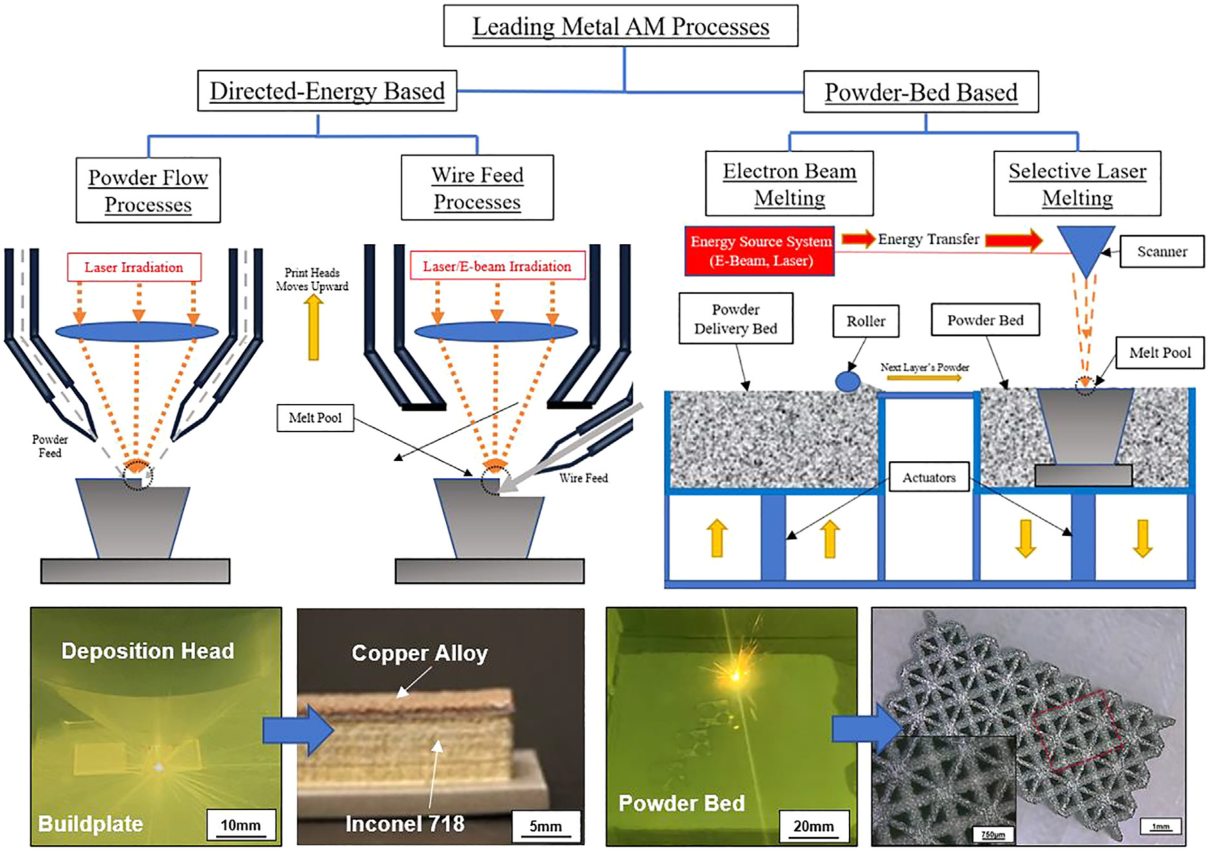

FIGURE 3

Directed energy deposition (DED) and Powder bed fusion (PBF) based additive manufacturing methods. Printing schematics reporduced from [24], Copyright 2018, with permission from Elsevier. Copper alloy-Inconel 718 image reproduced from [6], Copyright 2018, with permission from Elsevier.

limits manufacturers when trying to obtain obscure alloying element materials and control the melt's final chemistry, even with tight control of the wire feed rate. However, a big challenge is increasing the effective resolution due to the wire thicknesses and distortion challenges due to high heat input that causes warpage and difficulty in maintaining tolerances [36,37].

Comparatively, PBF utilizes a laser or electron beam to fuse a layer of powder, typically between 30 and 100 μm, that has been spread across the surface of the build-plate (or previously fused layer), as shown schematically from left to right in Fig. 3. After a layer is fused, the underlying part is covered with an additional layer of powder, and melt-cast layers surround the part from the powders. This technique mandates that the feedstock material is of one alloy composition, atomized as a single alloy or mixed or alloyed *in situ*. Also, a large amount of powder is required as it must both be fused into the part and surround the part during processing and remain until the end of a build. The small layer thicknesses and feature resolution (100–150 μm) compared to DED lend themselves well to printing more complex components with materials that have been well-established industrially. Naturally, this process is more common for industrial complex components used in the aerospace and biomedical fields, among many others, and is ideal for alloy design situations where high quantities of powder are available or previous work has been performed on a smaller-scale DED system. Feedstock powders can be combined in elemental form or small additions of one alloying element to a pre-alloyed feedstock that has been atomized from a commercial alloy. In both cases, large amounts of powder are required for performing the alloy design studies. Fig. 4b outlines a small design of experiment (DOE) tailored to understand the effects of the main PBF processing parameters such as laser power, scanning speed, and hatch spacing, all of which contribute to the effective energy density of the process and can influence the final properties and characteristics of the as-built specimens. While this experiment is only possible with one composition, different works have explored how process parameters and composition can influence the microstructure and properties of alloys that are created *in situ* using the PBF process or with different blends, thereby demonstrating how PBF can be utilized as an alloy development tool in addition to DED, albeit with a different set of challenges [38–42].

Past and current work on alloy design via metal additive manufacturing

There are currently two veins of research within the AM space regarding alloy design and development. The first is converting traditional alloys into AM-acceptable alloys, and the other is the discovery of new alloys. A large portion of the AM community seeks to find process acceptance within the larger manufacturing industry. There are well-documented success cases for specific applications and demonstration purposes [14,19], but metal AM parts' have had limited success being incorporated into

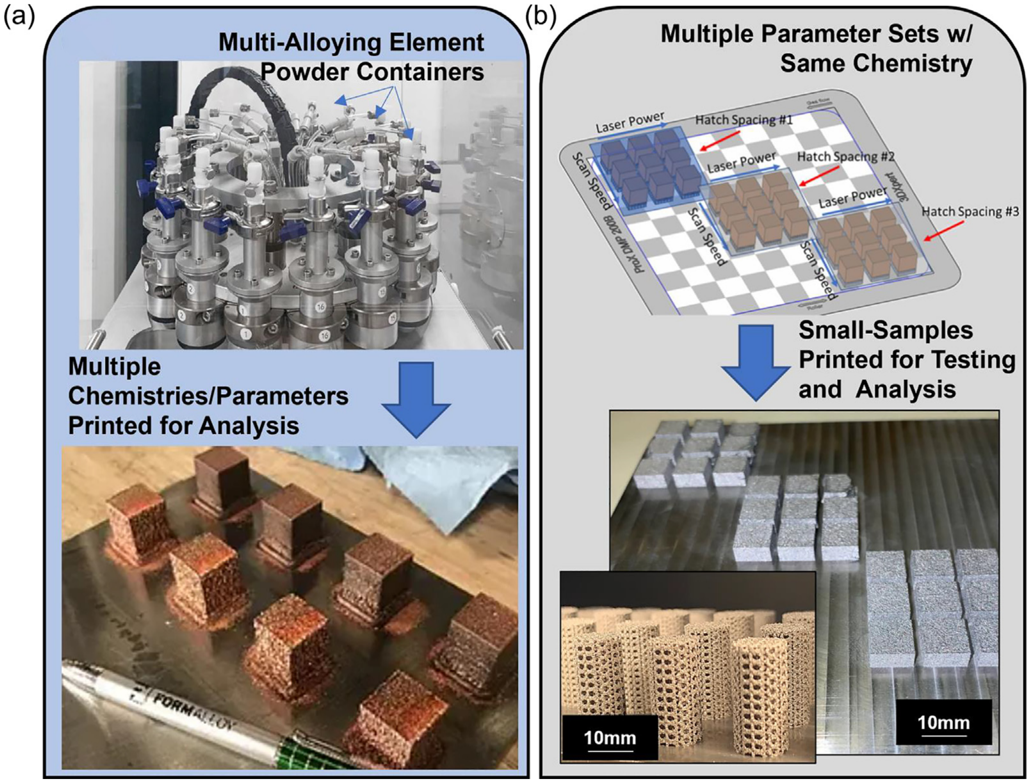

FIGURE 4

Examples of alloy design projects using both (a) the DED by leveraging multiple-powder feeder designs, and (b) the PBF technique by utilizing different parameter sets to yield variable microstructures and properties. All images provided by authors.

end-use products. In traditional manufacturing circles, the AM process itself is not proven robust enough for inclusion into end-use products. Compounding this issue is the limited information on AM alloy design-allowables and a general lack of material data necessary for certification of components in industrial applications. To this end, many resources have been devoted to developing and validating traditional, industry-proven alloys within the AM process paradigm instead of developing new alloys for emerging and exciting applications. Some alloy systems such as Ti6Al4V [43,44], Inconel 718 and 625 [45], and 316L SS [46] require no chemical alterations for AM processing, whereas aluminum alloy chemistries closer to eutectic compositions (AlSi10Mg and AlSi9Cu3) were required for AM processing to increase the wettability properties of the melt pool and decrease the shrinkage [47]. Where alloying elements are numerous, and the individual contribution of each element is vague, the practice of combinatorial guess-and-check methodologies and experimentation still yields positive results when basing alloys for AM from existing chemistries [48,49]. Combinatorial approaches for the processing of useful materials by metal-AM have been employed to synthesize magnetic materials [50], hydrogen storage alloys [51], high-entropy alloys [52,53], and bulk metallic glasses [54]. Most examples, however, are typically in the realm of trade secrets and IP within the industry. Despite this, significant strides in academia have been made in recent years towards utilizing AM as an alloy-design platform for emerging applications in the biomedical and aerospace industries, among many others. Studies typically combine theoretical and simulation of phase diagrams for a material system of interest, coupled with manufacturing experiments with the investigated alloy system and verification of the simulation results using subsequent mechanical testing and microstructural analysis. The most commonly used material systems range from nickel- and iron-based systems to aluminum- and titanium-based alloys or other magnetic materials and refractory high-entropy systems, investigated for next-generation applications.

An extensive amount of additive-based alloy design research has been devoted to iron- and nickel-based alloy systems that leverage complex combinations of reinforcing phases and microstructures, shown in Table 3. Dippo [32] demonstrated a well-integrated approach to alloy design of a modified Inconel 625 (Ni–Cr–Fr) chemistry using DED. Using the industrial alloy development feeder (ADF) from FormAlloy (see Fig. 4a), the solubility of alloying elements incorporated into Inconel 625 was able to be experimentally verified after CALPHAD simulations of precipitation. Further, the approach utilized can be scaled to analyze a significant amount of chemistries in a single day, with samples fabricated using the 5th (rotational) axis to print chemistries from one to the next to increase efficiency when performing electron microscopy and X-ray diffraction analysis. As mentioned previously, because of the nature of the “on-the-fly” elemental mixing, extensive optimization is required to understand the effects of argon gas flow rate and other operational parameters on the powder's mass flow rates, the melt pool, and the subsequent as-printed chemistries. Despite this challenge, a range of 0–1.7% difference in chemistry was observed

TABLE 3

Critical examples of alloy design approach leveraging metal-based AM.

Material System	AM Process & Objective	Key Results	Ref.
Inconel (Ni–Cr)–Nb	DED, increasing superalloy hardness	– Using powder blending and multi-alloy hopper via DED, good agreement between predicted phase information and as-printed phase composition can be achieved. – Presence of secondary reinforcing phases (Nb-based) can be predicted and confirmed using a CALPHAD-experimental approach.	[32]
Ni–Cr–B–Si	DED, Understanding Cr-Si-B effects on properties	– An abundance of hard Cr-rich precipitates produces high-hardness microstructures and can lead to cracking. – Scanning speed plays a major role in the cooling rate and subsequent crack formation in Cr-rich microstructures.	[54]
Ni–Cr–Si	DED + liquid droplet simulation, Understanding Cr-Si & cooling rate effects	– High Cr-Si content and rapid cooling rates result in metastable high-temperature silicides in the as-printed microstructures. – Small-scale arc-melting setup for creating controlled cooling rates can be used for further AM alloy design approach.	[55]
Aluminum 6061/7075 +Zr	PBF, alleviate cracking and porosity in the processing of Al alloys	– Application of an inoculant phase on metal powder feedstock can limit columnar grain growth during high-solidification rate additive processing. – Al_3Zr nucleant particles incorporated within Al 7075 feedstock enable its printability reliably with properties comparable to the wrought product.	[23]
Al–Ce	DED, Development of Al–Ce alloy processing range	– Highly variable microstructures observed near the melt pool indicate the variable growth velocities and thermal gradients within the melt pool during DED processing. – Good agreement of eutectic spacing and microstructure between thermodynamic modeling and experiment involving remelting of the as-cast plate in conditions representative of metal AM.	[56]
Ti–Cu	DED, elimination of columnar grains in AM Ti	– Copper addition to titanium to increase heterogeneous nucleation, thus forcing an equiaxed microstructure under high cooling rate processing. – Compositions ranging from 3.5Cu to 8.5Cu (wt%) enabled eutectoid microstructures that significantly affected both the strength (as high as 1023 MPa) and ductility (as high as 14.9%) of the alloy.	[57]
Ta–Ti	DED, Increasing bioactivity of titanium	– Authors demonstrated the enhanced biological response of titanium with the incorporation of tantalum without a decrease in processability due to tantalum's high melting temperature. – As low as 10 wt% tantalum was shown to have a strong biological response comparable to 100 wt% tantalum.	[27]

for each desired composition, including additions to Inconel 625 of Cr, Nb, Mo, indicating this approach's efficacy in designing and developing new alloys for advanced applications. Further, Hemmati et al. [54] studied the composition effects of variable Cr-B-Si amounts in Ni-based alloy on the microstructures and properties when processed via LAM. While no computation work was performed, the authors found that high Cr content added and a low Si-to-B ratio tended to have a harmful increase in crack pathways in the microstructure. Interestingly, the authors found that scanning speed played a critical role in crack formation due to the change in effective cooling rate (see Fig. 5a). In a related study, Li et al. [55] investigated compositional effects of Ni–Cr–Si alloys using a simulated high-cooling-rate processing setup. Thermo-Calc software was utilized to predict the Ni-rich corner of the Ni–Cr–Si ternary system to identify compositions likely to result in silicide formation (for wear resistance) and sufficiently high Cr-content for oxidation resistance. By combining a computational-experimental approach, the authors demonstrated that the cooling rate could play a significant role in the phases that form in the microstructure, i.e., high Cr and Si compositions tended to retain high-temperature (metastable) silicides in the microstructure due to the rapid cooling rate. Interestingly, the authors demonstrated that small-scale droplet experiments via arc-melting apparatuses at controlled cooling rates closely resemble LAM experiments. Such a result is significant as it validates that the alloy development approach using LAM can yield similar results to conventional approaches (see Fig. 5b). Other work has involved using chemical gradients to design structures with site-specific alloy compositions [22].

Among other material systems that have seen significant interest in using AM as a design platform are aluminum- and titanium-based systems [56–63]. Interest in processing by AM aluminum alloys with non-eutectic compositions has significantly emerged to improve the properties and consistency of parts produced via AM. Although it is known that aluminum alloys are typically easily machinable, the added benefits of utilizing AM have greatly influenced research into reliably fabricating components using this method. Martin et al. demonstrated the use of nano-functionalization whereby small powders are mixed with the parent aluminum alloy material to control solidification (see Fig. 1) [23]. The nanoparticles' presence in the microstructures prevents the average columnar grain growth and promotes a refined, crack-free, equiaxed grain structure. The nano-functionalized material's strength and ductility were

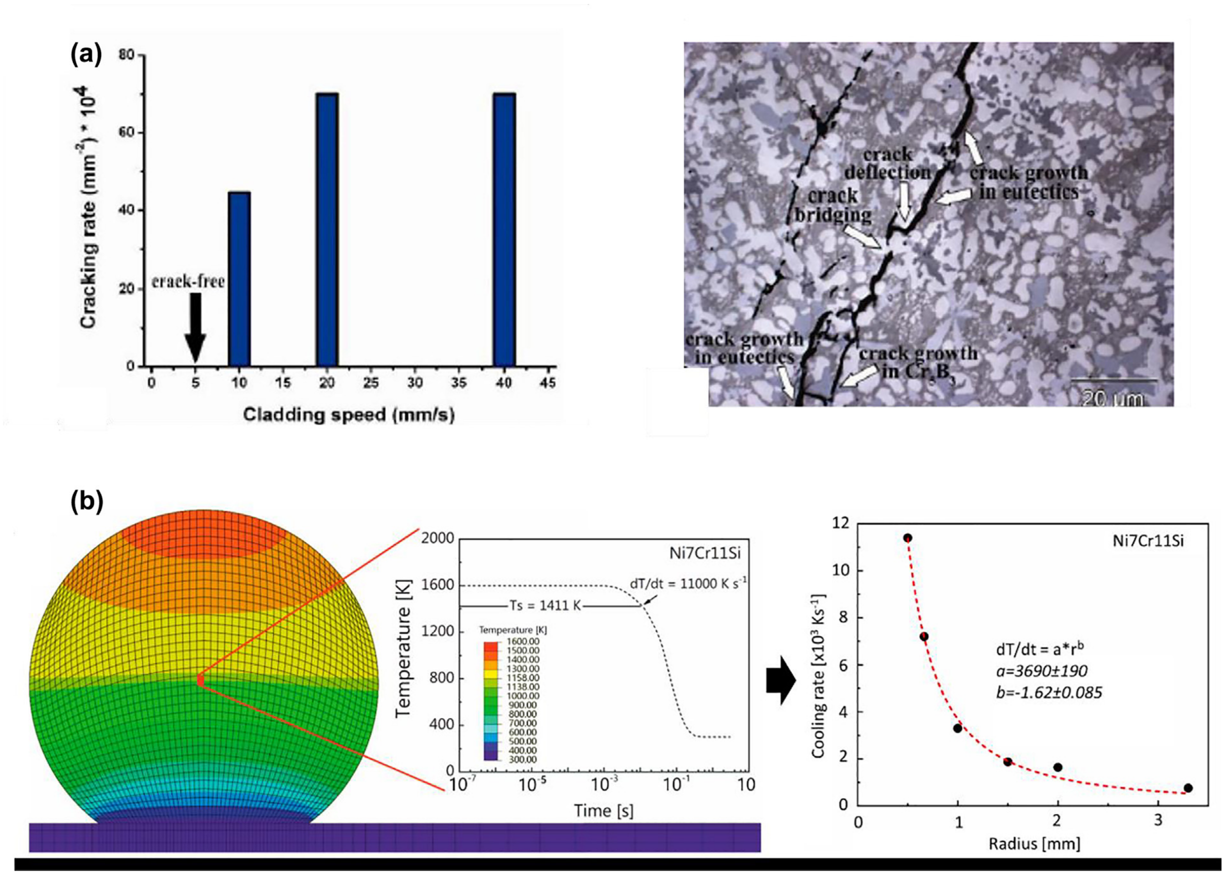

FIGURE 5

Examples of alloy design characteristics of Fe- and Ni-based alloys. (a) Example of scanning speed effects on microcracking in Cr–Si–B modified Ni-based alloy, reprinted from [54], Copyright 2013, with permission from Elsevier. (b) Example of a droplet-based simulation to understand the effects of cooling rate on the resulting microstructures of Ni–Cr–Si ternary system during laser-based AM, reprinted from [51], under Creative Commons CC BY License.

superior to both the base material (Al 7075) and a standard cast alloy that has been printed extensively (AlSi10Mg). Similar approaches have also been reported to modify other aluminum alloys for AM [49,64–68], with an example of the grain refinement mechanism shown in Fig. 6a. In another Al-related work, Plotkowski et al. [56] demonstrated the use of thermodynamic modeling to develop a processing range for Al–Ce alloys using a combination of eutectic models of dendritic and cellular growth given a specific set of processing characteristics. By laser melting various as-cast Al–Ce alloy plates, the authors developed experimental relationships between composition and microstructure in the as-fused region, which allowed them to model a processing range for various Al–Ce alloy compositions and interface velocities (indicative of processing conditions) (see Fig. 6b). A good qualitative agreement was observed for the eutectic spacing and general microstructure characteristics, given a specific set of processing qualities, providing manufacturers an understanding of the relationships between input parameters and resulting microstructure and properties for a new Al–Ce class of alloys. Further work with an Al–10 wt% Ce alloy composition processed via PBF resulted in high relative density composites with improved properties over cast counterparts. In an interesting wire-arc additive manufacturing (WWAM) study, Shen et al. [69] utilized a multi-wire system to develop Fe_3Al material by adjusting the feed rates of Fe and Al wires to achieve the desired 25 at% Al content. The authors demonstrated that this approach could achieve adequate compositional homogeneity despite working from the elemental feedstocks, substantiating this alloy design method using well-established rod-like feedstock materials.

Significant work has been performed on titanium alloys to understand and reduce columnar grain formation tendency during processing. Zhang et al. [57] demonstrated that copper addition to titanium via *in situ* alloying reduces the tendency for columnar grain formation due to high constitutional changes supercooling capacity that causes heterogeneous nucleation and a columnar to equiaxed grain structure transition during layer-by-layer processing. Copper's high diffusion rate in titanium at compositions ranging from 3.5 wt% Cu to 8.5 wt% Cu enables eutectoid microstructures that significantly affect both the strength (as high as 1023 MPa) and ductility (as high as 14.9%) of the alloy (see Fig. 7a). Other unique approaches towards alloy design incorporate various methods and alterations to existing materials and AM processes to achieve particular goals in such alloy systems.

An approach from Todaro et al. and Yuan et al. [70,71] utilized high-intensity acoustic vibrations to break up the grain structure within the melt pool, i.e., stimulating heterogeneous nucleation

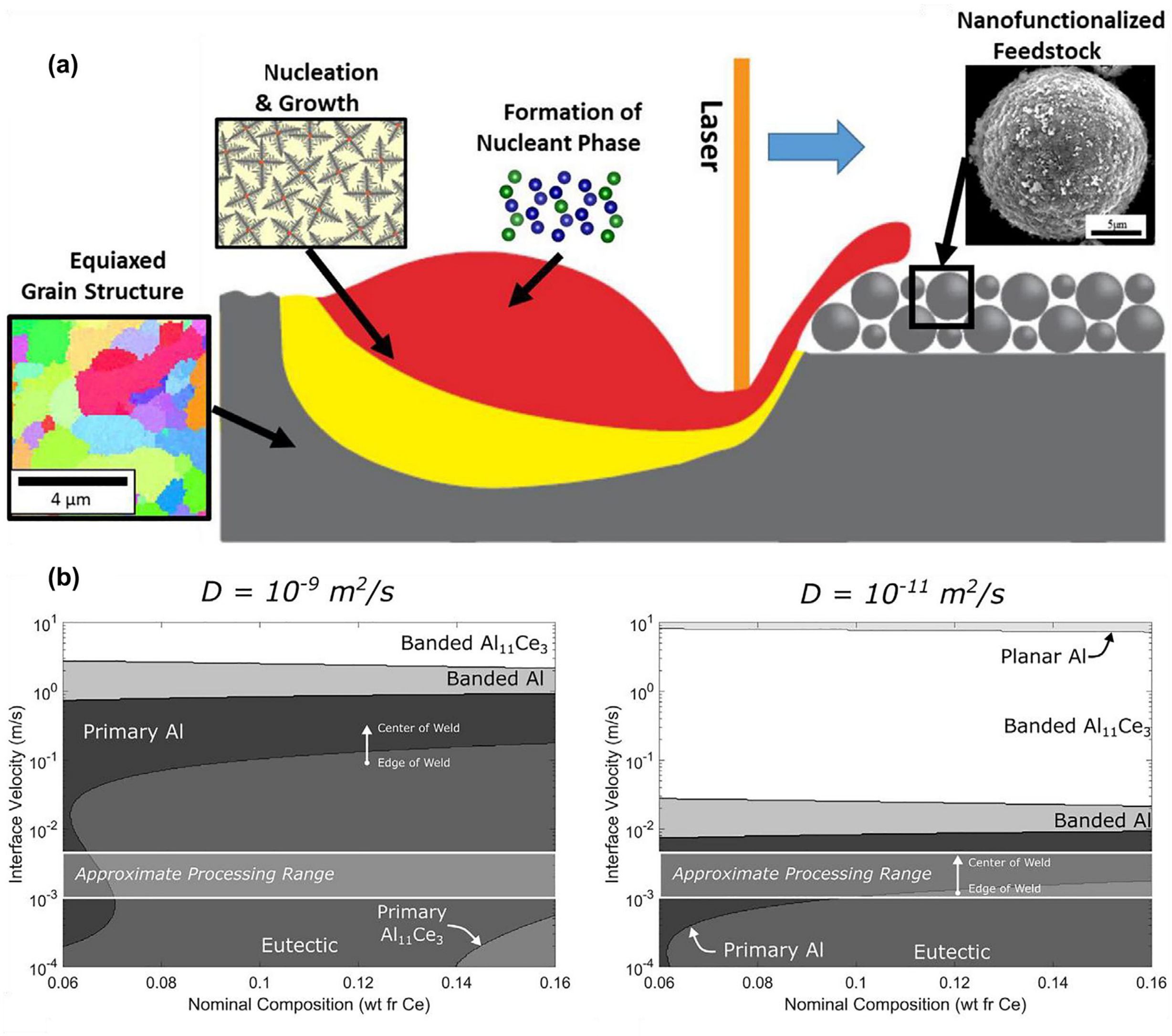

FIGURE 6

Examples of alloy design are being utilized in the development of aluminum-based materials. (a) Schematic showing the influence of the inoculant phase on the resulting equiaxed microstructure after solidification, reprinted from [64], Copyright 2020, with permission from Elsevier. (b) Resulting microstructures projected via computation coupled with experiments in the development of Al–Ce class of alloys, reprinted from [56], Copyright 2017, with permission from Elsevier.

and the formation of equiaxed grain structures (see the comparison of grain structure with/without acoustic vibrations in Fig. 7b). The authors demonstrated that acoustic waves cause significant "acoustic cavitation" in the melt pool by setting up an acoustic transmitter, which significantly agitates the molten pool and forces nuclei activation to promote fine equiaxed grain structures. Such an approach has the advantage of fabricating refined grain structures without inoculants or additives but does require an advanced setup that may be challenging for PBF-based methods as the current work was accomplished using DED. A different strategy for the same end-result in WAAM methods involves interpass rolling (i.e., mechanical work) applied between deposition layers to break up grain structures and promote finer microstructures than the standard columnar grain structures exhibited in this method [72,73]. Other works have focused on developing high-temperature intermetallic titanium aluminide structures and other *in situ* alloying via premixing of elemental constituents [74,75]. An interesting set of works from Mitra et al. and Bandyopadhyay et al. [27,76] investigated the incorporation of tantalum into titanium via DED to simultaneously increase the biocompatibility of titanium while also alleviating the processing challenges of tantalum, combining both *in situ* alloying and additional surface modification (see Fig. 8a). The authors found that as low as 10 wt% tantalum could be incorporated into titanium to significantly increase biological response, reducing the necessity for the refractory alloy that is ever-challenging to process and typically remains in powder form in the microstructure within some regions. In another wire-based AM work, the authors utilized a "combined cable" (see Fig. 8b) approach to fabricate high-entropy alloys with as many as seven constituents in a single pass [77]. The authors commented that a slight reduction in aluminum content compared to the desired amount was due to splashing in the melt pool, but compositions generally represented the desired amounts.

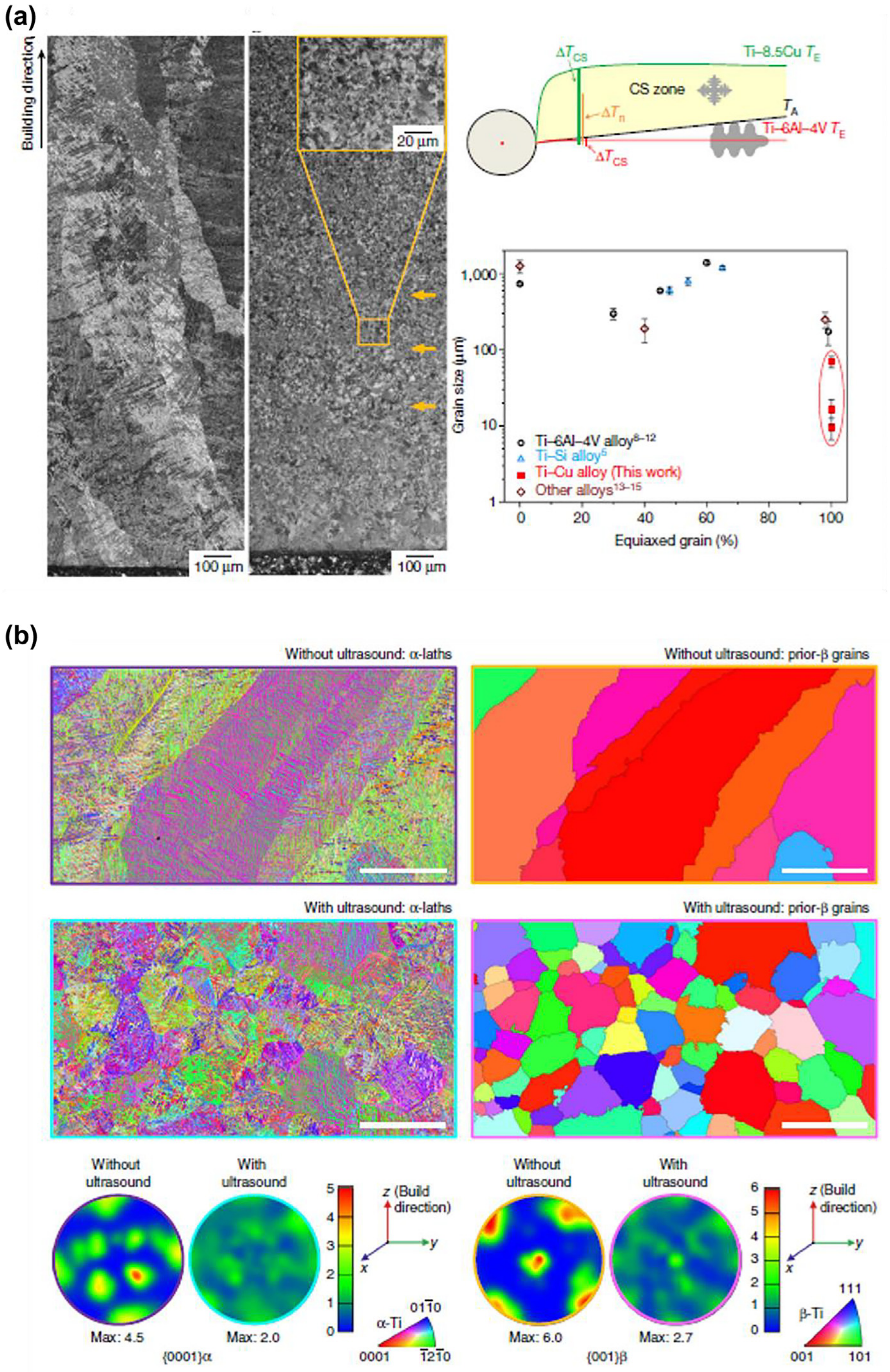

FIGURE 7

Examples of titanium-based alloy development via AM. (a) Copper-modified titanium influence a columnar to equiaxed grain structure, reprinted by permission from Springer Nature Customer Service Centre GmbH: Springer Nature, [57], Copyright 2019. (b) Effect of ultrasonic vibrations on grain refinement and reduction of columnar structures in DED-based processing of titanium alloys showing absence of texture and equiaxed microstructure, reprinted from [70], under Creative Commons CC BY License.

In metal AM another exciting alloy and material development area is developing metal–ceramic composite materials and structures [78–84]. While not specifically alloy design due to the nature of the composite materials, the strategies for creating these structures using AM greatly complement the recent developments in alloy design strategies using metallic materials' ductility, thermal/electrical properties, and a wide range of processability, with a ceramic material's hardness, strength, electrical and other properties to create structures with a variety of (in the best-case scenario) tunable properties. These structures are typically processed using powder-based methods with the ceramic powder premixed in anywhere from 1 to 20 wt% [85], but can also be processed in a coating form on top of an existing metallic structure to bring higher wear resistance and/or desir-

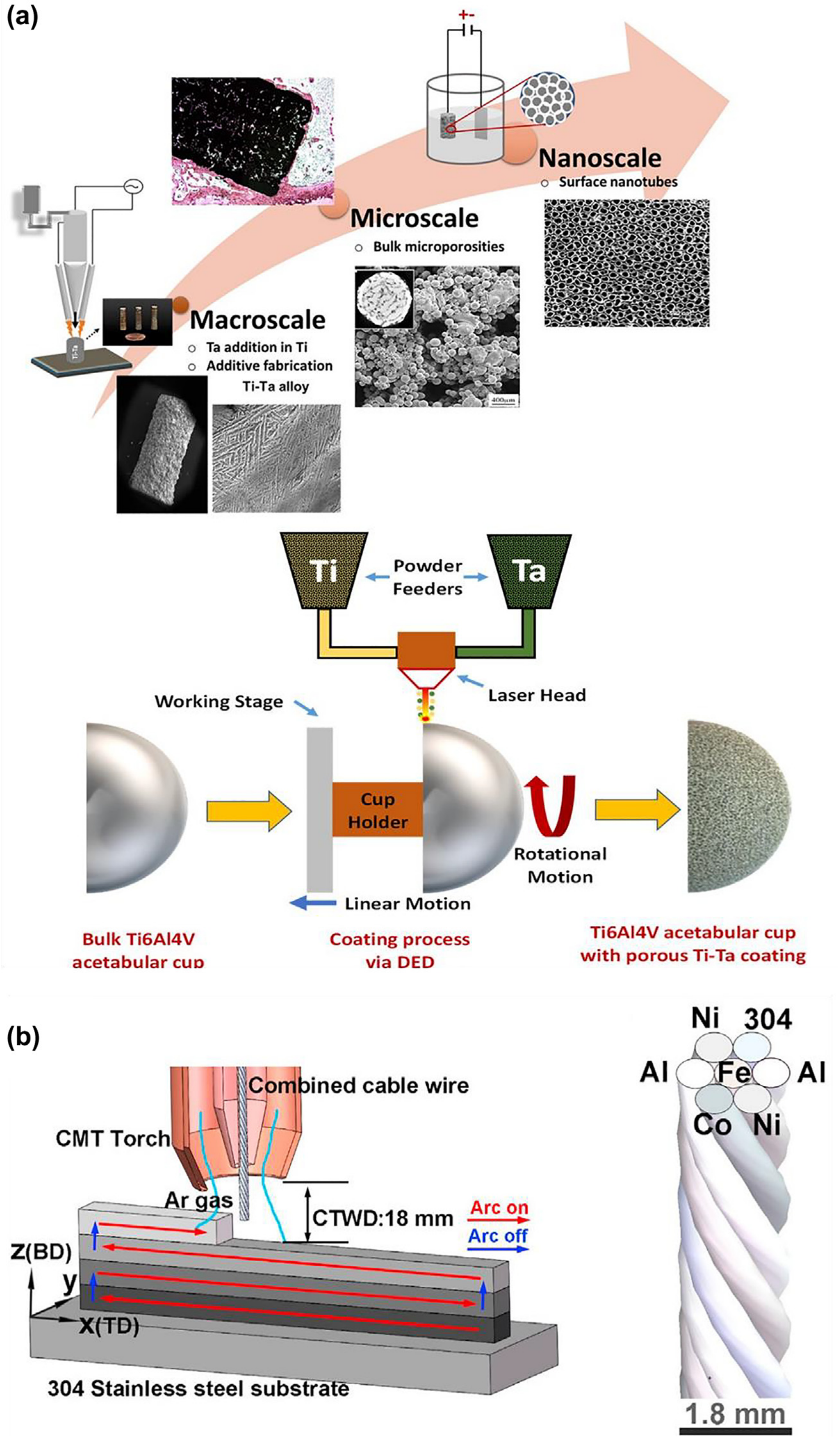

FIGURE 8

Novel multi-material concepts in alloy design using AM. (a) Application of DED to design Ti–Ta alloys to increase biocompatibility and balance processability challenges of incorporating tantalum in the microstructure, reprinted from [27], Copyright 2021, with permission from Elsevier (b) Combined cable concept for multi-material AM using wire-based DED, reprinted from [77], Copyright 2021, with permission from Elsevier.

able properties to the underlying structure [86,87]. Among various studies, Traxel et al. [79] have investigated BN and B_4C reinforcement to simultaneously improve titanium matrix composites' mechanical and oxidative properties to increase the possible service temperature. The authors found significant *in situ* reactivity between the titanium matrix and the reinforcement particles that formed new phases during the rapid solidification process that improved properties compared to titanium. In other material systems, Cooper et al. [88] investigated Inconel 625 reinforced with silicon carbide, aluminum oxide, and titanium carbide to manufacture a superalloy with improved temperature properties without high machining expenditure costs associated with superalloys owing to AM's ability to reduce material usage. The authors found that titanium carbide reinforcement significantly increased the microhardness while not limiting the processability due to cracking using other reinforcement phases. Plasma-transfer arc welding has also been utilized to fabricate these composites by incorporating a ceramic within the deposited powder mixture [89]. The authors showed good mechanical properties, high hardness, and low porosity, indicating its efficacy in forming metal matrix composites (MMCs) in a tool-less fashion. These approaches highlight how ceramic reinforcement can be utilized in small amounts to significantly influence the properties of materials produced using AM.

While premixed composites have seen extensive work, other research has shown the ability to transition from processing metallic-based materials to ceramics for increased surface hardness, biocompatibility, and thermal stability, among other desirable characteristics in different applications. Sahasrabudhe et al. [90] investigated laser surface remelting of a titanium substrate in a nitrogen-rich atmosphere to form a hard nitride phase near the surface. This demonstrated that a laser-based setup could be utilized to form a nitride-rich layer on any surface of an as-printed component to influence the wear performance and surface compatibility in different applications. In another work, Gualtieri and Bandyopadhyay [86] demonstrated that superhard ceramic vanadium carbide could be processed on top of a stainless steel substrate by utilizing a compositional gradient from the stainless steel to the ceramic. This work demonstrated the ability to transition using this technology in the development of site-specific alloying and reinforcement.

Current challenges and future trends

The motivation for innovating new alloys is based on the hypothesis of exceeding the performance limitations of pure metals in engineering applications. The widespread use of metal AM in various demanding applications, high cooling rates, and non-equilibrium processing strategy warrant designing new alloys specifically for AM rather than the legacy alloys borrowed from conventional manufacturing. Renewed interest in alloy design via AM is happening worldwide for metal-AM operations. For example, do we need Ti6Al4V or Ti5Al3.5V as a better option in metal–AM processing? Or do we need another alloying element to stabilize the beta-Ti phase further to maintain higher fatigue resistance in AM processed parts inherent in Ti6Al4V alloy processed via conventional approaches? It is anticipated that innovation in alloy design will shape the subsequent decades of manufacturing using metal AM. Since legacy alloys have been used in various metal AM operations for the past three decades without any composition modifications, it is anticipated that new alloys designed for AM operations will also be translated to conventional manufacturing, such as different casting and forming operations. This review summarized current efforts in alloy design via AM; however, this section explicitly addresses current challenges, future trends, further elaborating differences in strategy between conventional and AM-based alloy design.

While there has been significant investment in developing new alloys with metal–AM using DED and PBF, specific challenges exist that continue to limit the widespread adoption of the methodology. Because of the complex metallurgical phenomena during the high cooling rate processing [90], physics-based simulation, and new simulation methods' derivation, new alloys form a large part of the subject area [91,92]. As manufacturers continue to adopt AM technology, infrastructure will need to accommodate new machines and powder/wire inventory, similar to that used for production, for alloy design, and experiment with how they fit into the broader workflow and system utilized by the manufacturer. Critical differences between lab-scale AM material development and full-scale production must be addressed, and looking at existing technologies can often shed light on problems faced in alloy design using AM. Further, the process's complexity and components challenge many standard non-destructive evaluation techniques and open doors to new *in situ* monitoring and build-quality evaluation methods [94–96]. In process-based simulation, there are many reviews [93,97–99,105–107] available within the AM literature. However, these simulations require density functional theory (DFT) calculations for atomic positioning in broad strokes, which typically lends to the phase diagram's Monte Carlo simulation. Transformation-based and continuous-cooling/heating diagrams all then supplement the phase diagram. Physical experimentation is the last step to validate simulation predictions. The current cutting edge of alloy and material discovery employs machine learning (ML) as part of the simulation process. ML becomes a similar but more systematized version of the previously mentioned combinatorial guess-and-check practice as part of the discovery loop. Many researchers are currently investigating ML's use and have had some successes in predicting porosity and process maps for determining processing parameters [99,100], a typically time-consuming process when performed experimentally. Improvements in these areas are envisioned to significantly increase alloy design strategies using LAM to reduce entry's financial and intellectual borders.

An emerging area that is increasing attention towards future development is processing functionally/structurally-graded components using either bimetals or metal–ceramic composite combinations (see Fig. 9) [101–103]. These structures combine the best of multiple materials by varying the composition and structure of a component within a single part, sometimes incorporating multiple metals or ceramics for site-specific properties. Because of the multi-material nature of these structures, DED is the prevailing technology for their development. Examples of some of these structures are shown in Fig. 9, with functional transitions from immiscible/incompatible/challenging combinations of aluminum and stainless steel to titanium (Fig. 9a and b) [28,30], Copper-Inconel (Fig. 9c) [31], and magnetic–nonmag netic/other steels (Fig. 9d) [101], among many others. Because of these structures' variable properties, extensive thermodynamic and modeling work is emerging to help increase process reliability and reduce the required trial-and-error in creating such structures, namely developing compositional gradients that avoid the formation of brittle intermetallic phases and microstructures [104].

Moreover, unique structures composed of metal-ceramic interlayers (Fig. 10a) based on naturally-occurring structural armor in nacreous creatures and bone can be envisioned and created, providing site-specific properties to nearly impossible struc-

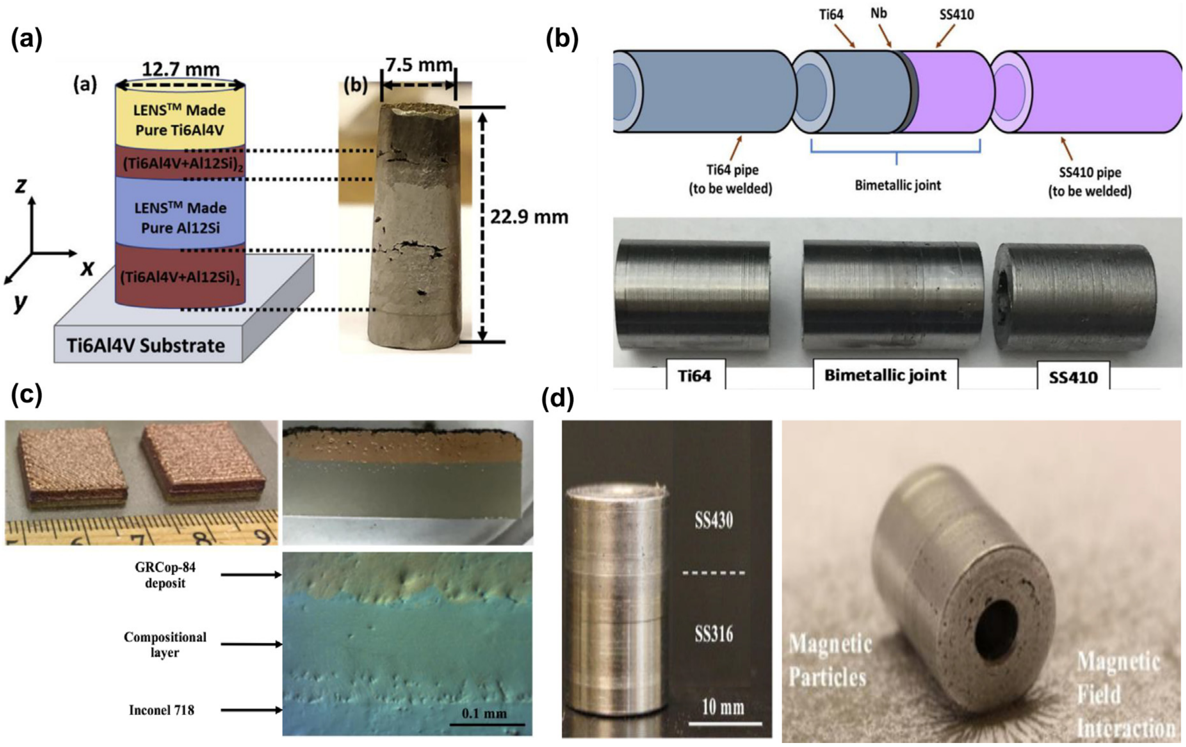

FIGURE 9

Examples of next-generation functionally-graded materials (FGMs) and related efforts towards achieving such structures with variable processing strategies. (a) Processing of Ti–6Al–4V/Al12Si joints, reprinted from [28], Copyright 2019, with permission from Elsevier. (b) Bimetallic structures fabricated from Ti–6Al–4V and SS410, reprinted from [30], Copyright 2020, with permission from Elsevier. (c) Increased diffusivity from deposition of copper alloy on Inconel 718, reprinted from [31], Copyright 2018, with permission from Elsevier. Magnetic–non magnetic joints are made from different stainless steel reprinted from [102], Copyright 2018, with permission from Elsevier.

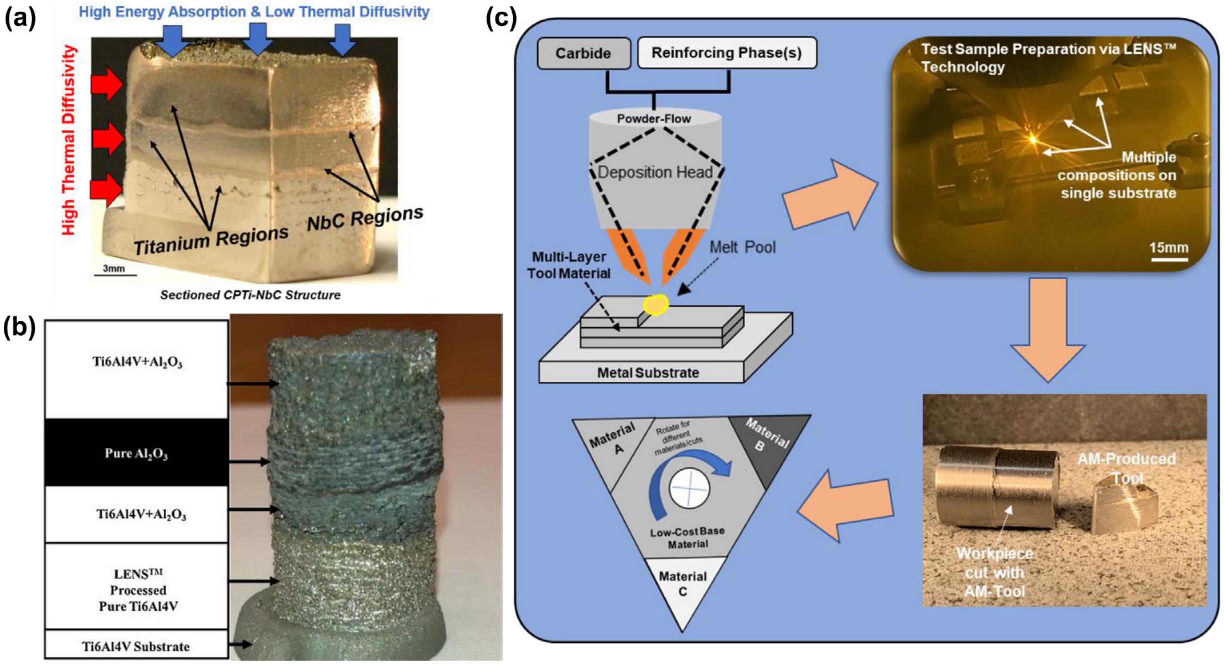

FIGURE 10

Examples of metal–ceramic composite material development. (a) Layered metal–ceramic composite, reprinted from [84], Copyright 2020, with permission from Elsevier. (b) Titanium to alumina (Al_2O_3) structure, reprinted from [101], Copyright 2018, with permission from Elsevier. (c) Diamond reinforced cutting tool design using DED, reprinted from [85], Copyright 2021, with permission from Elsevier.

tures to manufacture with traditional manufacturing methods [84]. LAM also fabricated structures of titanium–alumina composites and diamond reinforced cutting tools [101]. These examples show the efficacy of LAM to manufacture composite materials. Due to the demand for these structures and manufacturing capability, FormAlloy developed the Alloy Development Feeder (ADF) in 2019. Sixteen different compositions can be deposited using the ADF and its revolver-style hopper system to prevent cross-contamination between materials. With ADF, a single feeder can deposit new alloys or manufacture the most complex FGMs imaginable. With the intelligent design of these structures, the need for advanced alloys with multiple rare-earth additions may not be necessary if components can combine the best of multiple standard materials in single components, significantly alleviating concerns for sourcing of rare-earth elements needed in the most demanding alloy applications.

Applying either ceramic or metallic coating on powders for AM, thus forming functional core–shell powders, is another novel approach in the AM arena that holds great promise for AM of smart composite materials. The shell may be used as a thermal barrier to prevent melting by the energy source and related mixing, diffusion barrier, reflective/absorptive surface, enhance metallurgical bonding between the matrix and the reinforcement, enhance properties of the end material, etc. [108]. Metal-ceramic composite coatings for biomedical applications are another area of new materials development using AM. Either premix powders or on-demand deposition of various ceramics with metal powders can create composite coatings on metallic implants with enhanced biocompatibility or improved wear resistance for load-bearing applications [109–111].

Despite the increasing opportunities for alloy design in the materials engineering field using LAM, challenges still exist that need to be addressed to achieve full utilization. For some applications, these challenges are mainly technical and rely on aspects such as the high-cooling rate nature of AM and material compatibility, which pose a significant challenge in the processing reliability of some of the primary metallic materials used in AM. However, for other applications, material availability in powder or wire feedstock can lead to process-chain challenges in application spaces that have not caught up with the demand for alloy design in AM yet. These challenges ultimately are met with the age-old questions of property reliability and confidence, which can only be alleviated with extensive testing and characterization performed in the lab. We envision that, just like the development of AM from its infancy to now, alloy design using AM will see a similar growth pattern. Initially, AM users were using the technology to create prototype models for “touch and feel” purposes, while those currently applying alloy design concepts have been mainly limited to small-scale setups with small batches of powders and have not found the right combination of need, availability, and performance to warrant larger-scale studies or implementation. However, just as the continued push of technology development in AM has led to increased investment and implementation, so will technology development push those who wish to leverage this technology to develop new alloys and composites to meet the needs of the next generation of applications and performance.

Summary

Additive manufacturing (AM) has rapidly changed the landscape of large- and small-scale production environments across many industries and opened up opportunities for re-envisioning alloy design for emerging applications. With the onset of directed energy deposition (DED) and powder bed fusion (PBF) additive-based processing, new alloys can be designed and evaluated rapidly at a lower cost than traditional methods. This work outlined the methods and mechanisms by which alloy design can be achieved using laser AM (LAM), namely DED and PBF in premixed and on-the-fly methods. Further, a discussion was provided on the advantages and challenges of using different methods and the different material systems and strategies found industrially and, in the literature, such as nickel, titanium, and aluminum alloys. Finally, a discussion was provided on future challenges and emerging trends such as simulation to increase processing reliability and develop functionally gradient materials and structures for specific applications. It is envisioned that both industry and academic researchers spearhead future alloy-design efforts leveraging LAM benefits in many applications.

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.

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Data availability statement

All raw data for this study has been presented in this manuscript.

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