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Carbon nanotubes as a unique agent to fabricate nanoceramic/metal composite powders for additive manufacturing

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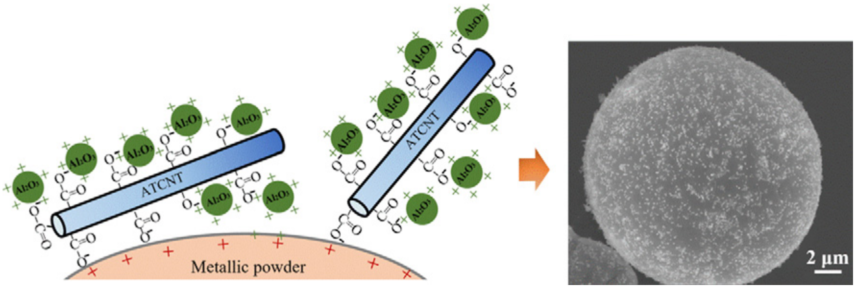
HIGHLIGHTS

- A novel application of CNTs was exploited for preparing nanoceramic/metal powders.
- The composite powders kept similar in shape, particle size, and distribution to raw metallic ones.
- The Al_2O_3-coated metallic powders showed higher laser absorptivity than uncoated ones.
- Al_2O_3 particles were uniformly dispersed and intimately contacted with the matrix after PBF.

GRAPHICAL ABSTRACT



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ABSTRACT

Laser powder bed fusion (PBF) offers many technological opportunities for producing high-performance composite parts with tailored structures. However, fabrication of suitable composite powders possessing homogenous dispersion, good flowability, suitable particle size and distribution is a prerequisite and main challenge currently faced. In this study, a novel strategy was developed to prepare nanoceramic/metal powders by using acid-treated carbon nanotubes (ATCNTs) as an agent. In detail, a 3 wt% ATCNT/Al_2O_3 colloid, in which the negatively-charged ATCNTs were partially covered with positively-charged Al_2O_3 nanoparticles under electrostatic attraction, was obtained by heteroagglomeration; subsequently, the uncovered surface areas of ATCNTs were intimately bonded to the positively-charged MoTiAl powders during their mixing. This ATCNT bridging made individual Al_2O_3 uniformly wrap on the surface of MoTiAl without aggregation. The Al_2O_3-coated MoTiAl powders remained similar in shape, particle size, and distribution to uncoated ones, simultaneously showing higher laser absorptivity due to an increased surface roughness. The PBF-processed Al_2O_3-ATCNT/MoTiAl composite was dense, in which Al_2O_3 nanoparticles were homogenously dispersed and intimately contacted with MoTiAl, giving rise to an increase in the hardness of the matrix.

1. Introduction

Owing to their high specific strength, sufficient stiffness and excellent wear resistance, metal matrix composites (MMCs) have received increasing attention in many applications including aircraft, automotive and energy storage [1,2]. In processing of MMCs, it is critical that the reinforcements are homogeneously dispersed and intimately incorporated into the matrix, which plays a vital role in determining the composite properties [3]. Currently, several manufacturing techniques such as casting [4], powder metallurgy [5,6] and mechanical alloying [7,8] are employed to produce MMCs. However, there are some existing problems, e.g., the insufficient densification rate and the irregular microstructure and aggregation of reinforcements, as well as the poor

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wettability and interfacial bonding between the reinforcements and the matrix, giving rise to resultant micro-cracks and even premature failure under loading [9,10]. In the meanwhile, the traditional methods usually involve highly time-, energy- and material-consuming processing steps [11]. Laser powder bed fusion (PBF), as a newly developed additive manufacturing technology, is capable of fabricating three-dimensional components directly by selectively melting raw powders using computer-designed CAD models [12–14]. Compared with traditional techniques, laser PBF provides many advantages, such as the direct production of complex structures, high material utilization rate, and high processing flexibility [14]. Furthermore, due to its non-equilibrium, rapid melting and solidification processing characteristics, laser PBF is expected to enhance the reinforcement/metal interfacial bonding [15]. Therefore, laser PBF offers new technological opportunities for producing high-performance MMC parts with tailored structures. Up till now, however, limited number of MMCs including TiC/TiAl [16], SiC/Fe [17], TiC/Al [18], TiB_2/Al [19],TiB_2/Inconel [20], Al_2O_3/Al [21,22], TiC/Ti [23], WC/Inconel [10], TiB_2/Ti [24], SiC/Al [25], TiB/Ti [26], TiB_2/TiAl [27], TiC/steel [28], $Al_2Si_4O_{10}$/Al [29] and Al_2O_3/Ni [30] by PBF have been reported.

The ever-increasing demand for novel heat-resistant materials beyond the realm of Ni-based superalloys has generated significant research interest in refractory intermetallics [31,32]. Mo-based alloys, possessing high melting points and high stiffness, are considered as promising candidates [32–34]. However, the unsatisfied elevated-temperature strength and poor processability of Mo-based alloys have seriously limited their application reliability [32,35]. It is well recognized that the mechanical strength of metals can be enhanced via the incorporation of uniform nanoceramic particles acting as a dislocation barrier [33]. Due to its low density, high hardness, high thermal stability, and excellent high-temperature creep resistance [36], the reinforcement of Al_2O_3 nanoparticle is widely used. For instance, Chen et al. [6] reported that Al_2O_3 dispersions could remarkably improve the elevated-temperature strength and fracture toughness of hot-pressed Mo_5Si_3. Therefore, PBF-processed Al_2O_3/Mo-based components with customized structures and enhanced performance have enormous potential for elevated-temperature service. Unfortunately, research regarding Mo-based alloys and their related composites by PBF, to the best of our knowledge, has rarely been documented.

It is known that the preparation of suitable powders is a prerequisite, but the main challenge currently faced is to broaden PBF in MMCs [14, 37]. The powders used for laser PBF should possess good flowability, suitable powder size (usually smaller than the diameter of a laser spot to be adequately melted), and narrow particle size distribution [38]. The previous studies [10,11,14–21,23,24] mainly focused on the morphologies and mechanical properties changed by reinforcements, while the influence of powder preparation on the PBF processability and final quality of MMCs was often ignored. In most works (see Table 1), the mixed powders were prepared by low-energy blending (LEB) and high-energy ball milling (HEBM). LEB is known for its feasibility of mixing two similarly micro-sized particles, while it is greatly limited for nanoceramic dispersion because nanoparticles are easily agglomerated when driven by strong van der Waals forces due to their high aspect ratio. In contrast, HEBM is capable of breaking tangled nanoparticles and dispersing nanoparticles into the matrix. However, HEBM involves uncertainty in tailoring the metallic powder size and distribution [11]. More importantly, severe plastic deformation usually occurs in HEBM-processed metallic powders, thus they tend to be flaky in the spherical shape or to be tangled with cold welding [16], showing poor flowability. Based on the above, it is imperative to develop a novel process that not only addresses the dispersion issue of nanoceramics but also keeps the features of the original metallic powders (e.g., shape, particle size, and distribution). This task is a dominating step for PBF.

The heteroagglomeration method is deemed to be an effective approach for mixing powders, such as in Al_2O_3/protein [39], SiO_2/polymer [40], and nano carbon/ceramic systems [41]. This method is based on the principle of electrostatic attraction that generally applies to two

Table 1
Characteristics of different methods in powder preparation for laser PBF.

Methods of powder preparation	Authors	PBF-processed MMCs	Starting powders and particle size	Advantages	Disadvantages
Low-energy blending (LEB)	Rong et al. [10,15]	WC_{1-x}/Inconel	WC_{1-x}: 15–45 μm, Inconel 718: 25–45 μm	Fast and easy; nearly keep the shape, particle size, and distribution of metallic powders; good powder flowability	Limited for similarly micro-sized particles; poor dispersion for nano-sized reinforcements
	Li et al. [25]	SiC/$Al_{12}Si$	SiC: 11–45 μm, $Al_{12}Si$: 20–63 μm		
	Attar et al. [26]	TiB/Ti	No information provided		
	Zhang et al. [20]	TiB_2/Inconel	TiB_2: 11–45 μm, Inconel 718: 5–12 μm		
High-energy ball milling (HEBM)	Han et al. [21]	Al_2O_3/Al	Al_2O_3: 10 nm, Al: 17 μm	Fast and easy; good dispersion of reinforcements	Involving uncertainty in tailoring the metallic powder size and distribution; metallic powder shape changed; poor powder flowability; including undesirable phases or contamination
	Jue et al. [22]	Al_2O_3/Al	Al_2O_3: 9 μm, Al: 1 μm		
	Gu et al. [23]	TiC/Ti	TiC: 50 nm, Ti: 23 μm		
	Gu et al. [18]	TiC/AlSiMg	TiC: 50 nm,$AlSi_{10}Mg$: 30 μm		
	Song et al. [17]	SiC/Fe	SiC: 27 μm, Fe: 20 μm		
	Li et al. [27]	TiB_2/TiAl	TiB_2: 3–5 μm, TiAl: 28 μm		
	AlMangour et al. [28]	TiC/steel	TiC: 50 nm or 1 μm, Stainless steel: 45 μm		
	He et al. [37]	TiC/Ti	TiC: 45 nm, Ti: 25 μm		
	Attar et al. [24]	TiB/Ti	TiB_2: 3.5–6 μm, Ti: 49 μm		
	Jue et al. [29]	$Al_2Si_4O_{10}$/Al	Al_2O_3: 9 μm, $AlSi_{10}Mg$: 30 μm		
	Gu et al. [16]	TiC/TiAl	Ti: 30 μm, Al: 16 μm, graphite: 30 μm		
Electro-codeposition	Ma et al. [30]	Al_2O_3/Ni, SiC/Ni	Ni, Al_2O_3: 50 nm, SiC: 50 nm	Good dispersion of reinforcements	Complex; costly, limited powder quantity
Melting + gas-atomization	Li et al. [19]	TiB_2/AlSiMg	nano-TiB2, Al—Si alloy, pure Mg	Good dispersion; spherical powder shape; good flowability; controllable size and distribution	Complex; costly; difficult for refractory alloys