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Single-layer graphene oxide reinforced metal matrix composites by laser sintering: Microstructure and mechanical property enhancement

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Abstract

This study investigates the integration of single-layer graphene oxide (GO) powders with iron matrix by laser sintering and its effects on mechanical strength and fatigue life. A laser-based additive manufacturing process is used to sinter GO and iron powders, and form GO–Fe nanocomposite materials. The aggregation of GO powders was prevented by the fast laser heating and cooling process. In addition, the evaporation of polyvinyl alcohol, which acted as a dispersing agent, from the cross-section helps align the GOs vertically in the cross-section. An energy-dispersive X-ray spectroscopy map from cross-sectional scanning electron microscopy images and Raman patterns together demonstrate the reduction of GOs after laser sintering. The GO–matrix interfacial structure was investigated by transmission electron microscopy. GOs were found to be stretched due to the rapid heating and cooling process during laser irradiation. Strengthening mechanisms of tensile strength and Young's modulus were developed based on the laser sintering results. Surface microhardness was increased by 93.5% by laser sintering of 2 wt.% GO. The improvement in the fatigue life after laser sintering of GO-reinforced iron matrix nanocomposites was also investigated.

Keywords: Laser sintering (LS); Single-layer graphene oxide; Metal matrix nanocomposites; Strengthening; Fatigue life

1. Introduction

Enhancing the strength of metals through microstructure design is very important for structural materials [1]. Controlling the microstructures, including grain boundaries, precipitates, dislocation forests or solute atoms, plays a key role in the design of materials with desired properties [2]. In general, microstructures that impede dislocation glide can lead to a significant increase in metal strength because plastic deformation mainly occurs via dislocation movement [3]. Embedding nanomaterials into a metal matrix to hinder dislocation sliding has attracted much attention in recent years.

Graphene oxide (GO) is an important material widely used to synthesize graphene and is considered as graphene functionalized by oxygen-containing groups [4,5]. Different research groups have contributed to revealing the functional groups of GO [6–10]. GO possesses some unique properties, which are distinctly different from those of graphene due to the existence of surface functional groups. GO has the potential for application in many fields, including microelectronic and chemical devices [11–13], energy storage [14,15] and composite materials [16]. The mechanical properties of graphene and GO have been investigated extensively by both experimental and theoretical approaches [4]. Experiments on bulk graphite yield [17] 1.02 ± 0.03 TPa for the in-plane Young's modulus. Lee

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et al. [18] used nanoindentation to measure the elastic properties and intrinsic breaking strength of free-standing monolayer graphene membranes; the measured Young's modulus was $E = 1.0 \pm 0.1$ TPa. Jiang et al. [19] investigated the Young's modulus of graphene through the intrinsic thermal vibration and molecular dynamics simulation, and found that the Young's modulus is in the range of 0.95–1.1 TPa at temperatures of between 100 and 500 K. Robinson et al. [20] measured the Young's modulus of reduced GO as 185 GPa through thermal annealing and analysis of the frequency response of suspended membranes. Suk et al. [4] found that the Young's modulus of graphene oxide relates to the number of layers: the measured Young's modulus of single-layer, two-layer and three-layer GO membranes were 223.9 ± 17.7, 444.8 ± 25.3 and 665.5 ± 34.6 GPa, respectively. These experimental results demonstrate the superb mechanical properties of graphene and GO.

GO has been used in polymer and metal matrix composites to enhance their mechanical properties. For instance, Bortz et al. [21] studied the fatigue life and fracture toughness by adding GO to epoxy matrix composites. It was found that the crack was pinned by GO, which helps increase the fatigue life of epoxy matrix composites. Wang et al. [22] determined that the tensile strength of GO-reinforced aluminum matrix composites was enhanced by 62% with 0.3 wt.% GO. However, manufacturing high-quality graphene-reinforced metal composite is still a challenge due to the aggregation of graphene nanoplatelets with current techniques and our lack of understanding of the GO–matrix interface. Recently, the interaction of graphene with metals and its effects on mechanical property has been studied. The strength of graphene or GO-reinforced metal composites was investigated by Wang et al. [22]. The enhanced strength of graphene/copper was discussed by Hwang et al. [23] and by Pavithra et al. [24]. The mechanism of dislocation blocking by graphene was investigated recently in Kim et al. [1]. However, there has been no systematic study of the mechanical properties of graphene–metal nanocomposite, especially fatigue performance. In this paper, we study laser sintering of single-layer GOs and iron powders to make GO–Fe nanocomposites with GOs uniformly distributed in the matrix. The fundamental mechanism underlying the enhancement in mechanical properties, i.e. elastic modulus, strength and fatigue life, will be investigated. The chemical distribution, interfacial structure of GO–Fe and the atomic structure of GO after laser sintering will be studied to help understand the mechanical property enhancement.

2. Experimental method

Iron powders were mixed with GO using polyvinyl alcohol (PVA) as a dispersing agent. AISI 4140 plate was chosen as the substrate. The samples were first austenitized at 850 °C for 20 min followed by oil quench and then tempered at 450 °C for 2 h and naturally cooled down to room temperature in a vacuum furnace. The iron powders (average diameter 4 μm) and single-layer GO (from Cheaptube Inc.) were laser sintered. The thickness of GOs is 0.7–1.2 nm (by atomic force microscopy) and the sizes of single-layer GO are in the range of 300 nm to several micrometers in XY dimensions. The concentration of GO in the Fe–GO nanocomposites was 2 wt.%. GOs were separated in the coated layer by PVA and randomly distributed over the cross-section.

An IPG fiber laser was used as a laser sintering energy source; it was operated with a laser power of 100 W and a frequency of 50 kHz. The pulse duration and wavelength were 220 ns and 1064 nm, respectively. The beam size is 0.8 mm, while the scanning speed and step size were 2 mm s^{-1} and 0.25 mm, respectively. Before laser sintering, the GO and iron powder were mixed together using a magnetic stirrer in water using PVA as a dispersing agent. PVA (4 wt.% of total solution) was used to separate GO [25,26]. After that, the mixed powders were coated on the sample surface. The substrate surface was mechanically polished with 0.05-μm grade aluminum oxide powder before coating [27–31].

Samples after laser sintering were prepared for Raman measurement. The surface and cross-sectional morphologies were characterized by a Hitachi S-4800 field emission scanning electron microscope and an FEI Philips XL-40 scanning electron microscope, respectively. A FEI Nova 200 focused ion beam (FIB) system was used to prepare transmission electron microscopy (TEM) samples by the lift-out method. The microstructure TEM images were obtained with an FEI Titan system operating at 300 keV.

Three-point bending fatigue test was performed in a 100 kN MTS servohydraulic fatigue test machine. In loading-control mode, a sine wave function with frequency of 10 Hz was loaded. The stress ratio R equals 0.1, where R is $\sigma_{min}/\sigma_{max}$ (σ_{min} and σ_{max} are minimum and maximum stress, respectively). The maximum bending stress is $\sigma = 3PL/2bh^2$, where P is the applied load, L is the span for the bending fatigue test, b is the width of the specimen and h is the thickness of the specimen. All the tests were carried out at room temperature.

3. Results and discussion

3.1. Cross-sectional morphology and chemical analysis after laser sintering

During laser irradiation, iron powders were melted into liquid and PVA was evaporated from iron liquid as bubbles. These PVA bubbles helped to align the GOs vertically in the cross-section of the laser-sintered layer. The schematics of the aligning process before and after laser sintering are shown in Fig. 1a and b. The cross-sectional microstructure of the laser sintered layer is shown in Fig. 1c. Energy-dispersive X-ray spectroscopy (EDS) maps of carbon and iron are shown in Fig. 1d and e, respectively. It can be clearly seen in Fig. 1d that the GOs are vertically aligned in the cross-section of the iron matrix. A thin layer