

Enhancement of the thermal conductivity of Cu Matrix by addition of RGO

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Abstract. Graphene have been condered as an ideal reinforcement material due to its fascinating physical and mechanical properties. In this study, the graphene reinforced the Cu matrix nanocomposites with excellent thermal conductivity have been successfully fabricated by hot pressing sintering of RGO/Cu nanocomposites powders which were prepared via situ chemical reduction method using NaBH₄ as reducing agent. The reduced graphene oxide and prepared nanocomposites were characterized by X-ray diffraction (XRD), Fieldemission scanning electron microscope (FESEM). The FESEM shows that the graphene homogeneous dispersion on the nanocomposites powders. Furthermore, the thermal conductivity of graphene reinforced Cu matrix has been improved compare with the Cu matrix without addition graphene.

Introduction

Graphene is a monolayer which composed of carbon atoms with sp²-bonded carbon tightly packed into a 2D honeycomb lattice[1-3]. This novel carbon material with its large surface area[4,5] and excellent thermal properties[6,7] has been considered as an ideal reinforcement material. Recent years, graphene reinforced composites has make a great progress[8], while the composites are mainly focused on graphene reinforced polymer composites, the strength and other properties of the polymer matrix were remarkably increased as the addition of graphene. However, the graphene reinforced metal matrix composites was rarely report[9]. Therefore, it is necessary to study the graphene reinforced metal matrix composites. On the other hands, the dispersibility of graphene play an important role in graphene reinforced metal matrix composites.

Currently, graphene can be prepared by various approaches, including micromechanical exfoliation of graphite[10], chemical vapor deposition[11] and epitaxial growth of graphene films[12] and chemical deoxygenation of graphene oxide[13], the chemical deoxygenation of graphene oxide is consider as the common way to obtain large-area graphene[14]. However, graphene sheets suffer from very poor dispersibility and irreversible agglomerates through π - π staking interaction between graphene sheets, which result it difficult to application in various areas. Then, great efforts are required to solve this issue. Recently, several ways have been proposed to obtain well-dispersed graphene solution. Zhang et al. reported that poly (diallyldimethylammonium chloride) (PDDA) acts as both a reducing agent and a stabilizer to prepare soluble graphene nanosheets from graphite oxide, the reduced graphene nanosheets exhibited single-layer structure and well dispersion in various solvents[15]. Zhou et al. developed a simple surfactant-free approach to dispersion graphene through a solvothermal reduction of graphene oxides in N, N-dimethylacetamide, and the as-prepared graphene could be re-dispersed well in more than six kinds of solvents[16].

In this study, RGO/Cu nanocomposites powders were fabricated through well-dispersed graphene solution via situ chemical reduction method using NaBH₄ as reducing agent. Furthermore, the graphene reinforced the Cu matrix nanocomposites were prepared by hot pressing sintering of

RGO/Cu nanocomposites powders. The microstructure and thermal conductivity of RGO/Cu nanocomposites were investigated by various analysis methods.

Experimental section

Materials

Flake graphite was purchased from Nanjing XFNANO Materials Tech Co, Ltd. dimethylformamide (DMF), copper sulfate (CuSO_4), sodium borohydride (NaBH_4) and other reagents were all obtained from Shanghai chemical Reagent Ltd.

Preparation of RGO/Cu

The reduced graphene oxide (RGO) reinforced Cu Matrix nanocomposites was obtained. Initially, graphene oxide (GO) was prepared based on the modified Hummers method[17], the well dispersion RGO was synthesized via 0.03g of GO dispersion in 150ml DMF solution then the mixture solution was transferred to an autoclave and heated to 180°C for 12h, Subsequently, the well dispersion RGO solution was added to the 450ml CuSO_4 solution (0.2 M) under the bath sonicator for 20 min at room temperature, when the well dispersion RGO solution was controlled to be 7.5 ml, 30 ml and 90 mL, the corresponding weight ratio of RGO to Cu in the RGO/Cu nanocomposite powders was 0.025 wt%, 0.1 wt% and 0.3 wt%, respectively. Followed the 350ml NaBH_4 solution (0.1M) drop into the above mixture solution by peristaltic pump. After finished the reaction, the products were filtered, washed, and vacuum freeze drying, the resulting samples were placed into the H_2 atmosphere with flow of 220 sccm under 250°C for 5h to remove the oxidized Cu. The RGO/Cu nanocomposite powders were compacted in powder pressing machine under 28 MPa, and the compacted powders were sintered by hot pressing at 900°C under 100MPa for 2h. The heating rate was maintained at $10^\circ\text{C}/\text{min}$ from room temperature to 850°C , and the heating rate was keep at $5^\circ\text{C}/\text{min}$ from 850°C to 900°C .

Characterization

XRD patterns were obtained on an X-ray diffractometer (RiGaKu, Japan) using $\text{Cu K}\alpha$ radiation. Morphological analysis was performed with a Quanta x50 fieldemission scanning electron microscope (FEI, USA) with an acceleration voltage of 20 kV. The thermal diffusivities were tested at room temperature by a laser flash apparatus (IFA 457, Netzsch).

Results and discussion

Fig.1 shows the XRD patterns of GO, 0.025Wt%RGO/Cu, 0.1Wt%RGO/Cu and 0.3Wt%RGO/Cu nanocomposites powders. It is obvious that the diffraction peak at 2θ values of 11.07° is ascribable to the GO characteristic peak[18], which is in accordance with previous reported. Then the RGO solution was synthesized when the GO dispersion in DMF solution and the mixture solution was heated to 180°C for 12h, After the NaBH_4 solution added to the mixed solution of RGO solution and CuSO_4 solution, the characteristic peak of the GO has disappeared and observed Cu and Cu_2O characteristic peak. It is suggesting that the GO has been reduced to RGO and the Cu^{2+} reduced to Cu nanoparticles in the presence of DMF and NaBH_4 , while the nanoparticles of Cu are oxidation to Cu_2O nanoparticles in the air. When the RGO/Cu nanocomposites powders were treated at H_2 atmosphere, the Cu_2O nanoparticles were reduced to Cu nanoparticles. This results can be confirmed by XRD patterns(Fig.2).

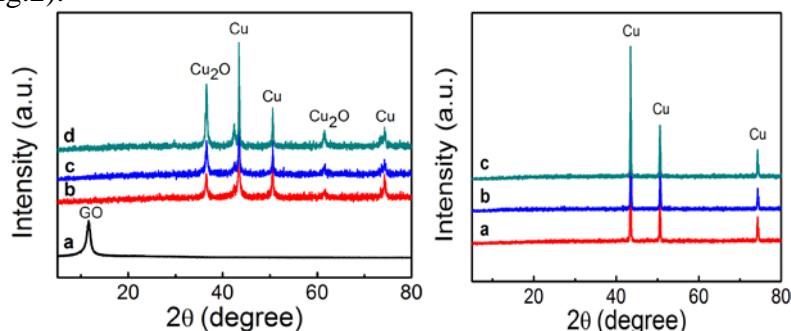


Fig.1 XRD of (a)GO; (b) 0.025Wt%RGO/Cu; (c) 0.1Wt%RGO/Cu; (d) 0.3Wt%RGO/Cu (left)

Fig.2 XRD of RGO/Cu after H₂ treatment (a) 0.025Wt%RGO/Cu; (b) 0.1Wt%RGO/Cu; (c) 0.3Wt%RGO/Cu (right)

Fig.3 shows a FESEM image of RGO/Cu nanocomposite powders with different RGO content. The pure Cu nanoparticles aggregate spontaneously and the size with diameter of Cu particles is in the range of 200-1000nm (Fig.3a). With the addition of RGO, the RGO layer prevent the agglomeration of Cu nanoparticles and was homogeneously covered by Cu nanoparticles, the Cu nanoparticles were ellipsoidal and about 500-1000nm in size (Fig.3b). This proves that no decomposition of RGO were made during the reduced process by H₂ and the Cu conformally wetted the surface of RGO. Furthermore, the FESEM of Fig.3c and Fig.3d revealed that the surface of RGO were covered with small Cu nanoparticles (about 20nm), this may attributed to Cu₂O nanoparticles were formed on GO were reduced to form Cu islands on the RGO layers.

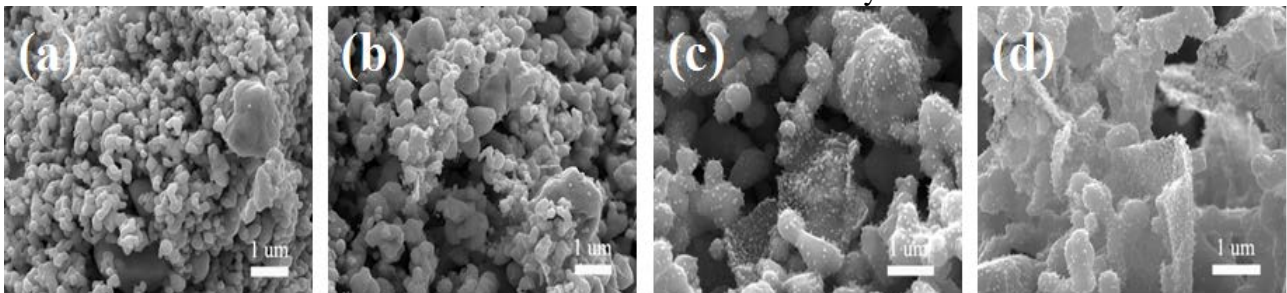


Fig.3 FESEM images of RGO/Cu nanocomposite powders with different RGO content (a) 0Wt%RGO/Cu; (b) 0.025Wt%RGO/Cu; (c) 0.1Wt%RGO/Cu; (d) 0.3Wt%RGO/Cu

RGO/Cu nanocomposites were densification by hot pressing at 900oC under 100MPa for 2h. Fig.4 shows the fracture surface FESEM images of RGO/Cu nanocomposite, the pure Cu has high density (Fig.4a) and the RGO layers were homogeneous dispersed in the Cu matrix (Fig.4b-d).

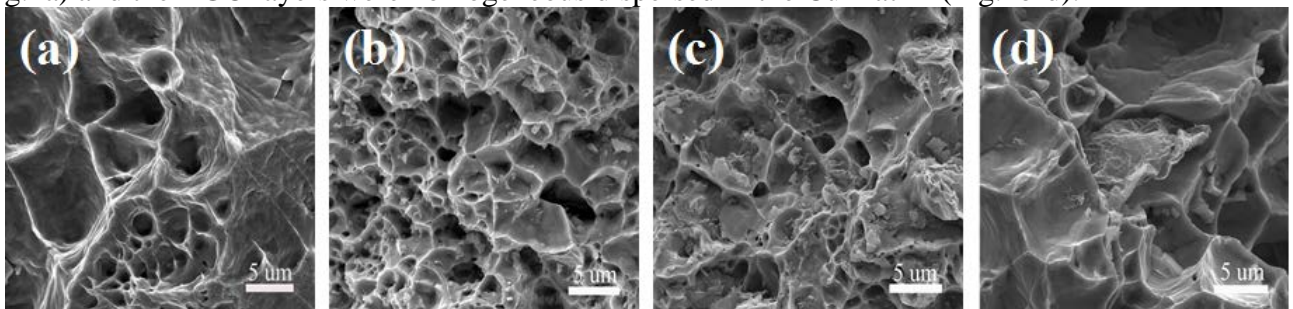


Fig.4 fracture surface FESEM images of RGO/Cu nanocomposite with different RGO content (a) 0Wt%RGO/Cu; (b) 0.025Wt%RGO/Cu; (c) 0.1Wt%RGO/Cu; (d) 0.3Wt%RGO/Cu

The measured thermal conductivities of RGO/Cu composites are shown by Fig.5, it is observed that the thermal conductivities of RGO/Cu composites are enhanced compared with pure Cu. This may attributed to graphene have an fascinating thermal conductivities, the RGO homogeneous dispersed in the Cu matrix and acts as reinforcements in Cu matrix.

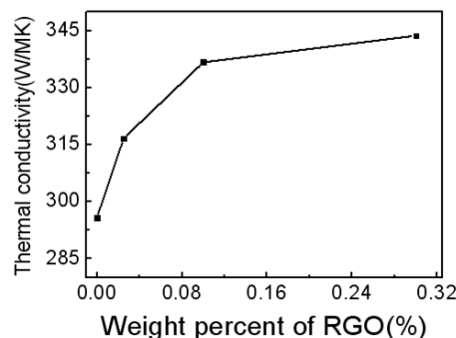


Fig.5 Thermal conductivities of RGO/Cu composites with different RGO content

Conclusions

In this paper, the RGO reinforced the Cu matrix nanocomposite was synthesized via chemical reduction and hot pressing method. The Cu₂O was effectively reduced to Cu nanoparticles by H₂, the RGO layer was homogeneously covered by Cu nanoparticles, which the size with diameter of Cu particles is about 20nm. The RGO layer homogenous dispersion in Cu matrix after hot pressing at 900°C under 100MPa for 2h. The thermal conductivities of RGO/Cu composites was increased compared with the pure Cu matrix.

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