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Preparation of nickel-coated graphite by electroless plating under mechanical or ultrasonic agitation

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ABSTRACT

In this paper, nickel-coated graphite powders were prepared by electroless plating. After a novel and facile pretreatment of graphite, electroless plating was carried out in an alkaline bath having nickel chloride ($NiCl_2$) as a source of nickel and sodium borohydride ($NaBH_4$) as a reducing agent. During the electroless nickel plating, mechanical or ultrasonic agitation was adopted to improve the properties of coating. The coated graphite powders were characterized by field emission scanning electron microscopy (FE-SEM), energy dispersive X-ray spectroscopy (EDX), and X-ray diffraction (XRD). The results show that two different morphologies of Ni–B coatings were achieved on the surface of graphite. Both coatings on graphite are continuous and uniform. For mechanical agitation process, the coating exhibits a sesame-seed slice-like structure which is composed of large spherical grains. The ultrasonic agitation process results in a faster deposition rate and a well-knit membrane-like structure coating free of voids. This is attributed to the easier nucleation and fine particle distribution of nickel nuclei under the ultrasonic agitation.

1. Introduction

As one kind of core–shell structure composites, nickel-coated graphite has attracted considerable interests in many areas, such as mechanical [1–4], electrochemistry [5] and electromagnetism [6,7] due to the good comprehensive properties. For example, nickel-coated graphite powders were usually added in metal matrix for production of engine axletree [4] and gas-turbine [8] because of the combination of good qualities of graphite and nickel/nickel alloy, such as unique solid lubrication, excellent corrosion resistance and high temperature resistance. The nickel-coated graphite powders can be produced by many methods, such as ion plating and chemical vapor deposition [1], electroless nickel plating [2,6,7], electroplating [9] and gas suspension coating [5]. Among these methods, electroless nickel plating is considered to be one of the most convenient and effective techniques [10–14].

It is well known that electroless plating is an autocatalytic process of depositing metals onto base material from aqueous solution without the application of an external electrical energy [15–19]. However, some base materials such as WC, SiC, Al_2O_3, carbon nanotubes, bamboo fabric and graphite, need to be pretreated in a sensitization solution and an activation solution because these materials do not have autocatalytic activity. This two-step sensitization–activation method in electroless plating has been widely reported [7,8,20–22]. Nevertheless, some complex technical problems still exist in the conventional sensitization and activation process [13]. In general, the activation solution contains noble metal salts. The cost of noble metal would limit the wide applications of noble metal activation. Recently, some studies have focused on electroless plating on non-metallic materials using simple and feasible pretreatment approaches. Palaniappa et al. [8] reported that the activation of graphite powders can be achieved by means of heat treatment in muffle furnace. Luo et al. [13,23] prepared the nickel-coated WC and nickel-coated Cr_3C_2 powders by ultrasonic-assisted electroless plating without employing conventional sensitization and activation. Deuis et al. [21] pointed out that nickel oxide activated layer on Al_2O_3 could be obtained through the decomposition of nickel nitrate by heat treatment. However, few investigations about electroless nickel plating on graphite without noble metal activation have been reported [8]. Furthermore, the effect of distinct agitation modes on the surface morphology of electroless nickel alloy coating has been limited studied.

In this paper, a simple and economical pretreatment without two-step sensitization and activation produce was carried out on graphite. Nickel-coated graphite powders were prepared in an alkaline bath that contains $NiCl_2$ and $NaBH_4$. The surface morphologies, phase composition and deposition rate of the Ni–B coatings were investigated in detail. The influence of agitation on the nucleation and growth mechanism of Ni–B coatings was also analyzed.

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2. Experimental procedure

2.1. Pretreatment of graphite powders

Graphite powders were supplied by Tianjin Kermel chemical reagents Co., Ltd. in the present study. Prior to electroless nickel plating, surface pretreatment of graphite powders was carried out. Firstly, graphite powders were degreased in acetone solution with ultrasonic cleaning for 30 min at 40 °C. Then the powders were stirred and heated at 60 °C in distilled water to remove the residual acetone. Secondly, the graphite powders were immersed in 20 wt.% hydrofluoric acid (HF) solution at room temperature with ultrasonic processing for 30 min. Then the powders were rinsed with distilled water several times until the pH value was 7. At last, the pretreated graphite powders were desiccated in a vacuum drying oven.

2.2. Electroless nickel plating

The pretreated graphite powders were introduced into a 500 ml beaker filled with 200 ml nickel bath. The nickel bath consisted of metallic main salt ($NiCl_2$•$6H_2O$), reducing agent ($NaBH_4$), complexing agent ($C_6H_5Na_3O_7$•$2H_2O$) and buffer agent (NaOH), as listed in Table 1. All the chemical reagents were analytical reagent grade. In this study, we synthesized nickel-coated graphite powders under two distinct agitation processes: mechanical agitation (the agitator is made up of polytetrafluoroethene (PTEF), agitating speed was 200 r/min, and agitating diameter was 5 cm), ultrasonic agitation (a constant ultrasonic frequency of 25 KHz with 250 W powers). The schematic diagram of experimental equipment is shown in Fig. 1. During the electroless nickel plating processes, the pH values of solutions remained 12 by 20 wt.% NaOH solution and the temperatures were maintained at 40 °C.

2.3. Characterization

Contact angle between graphite and distill water was measured at 25 °C using a JY-82 goniometer. Surface morphologies and chemical composition of coated graphite powders were characterized by field emission scanning electron microscopy (FE-SEM, 4800S, Hitachi, Japan) coupled with an energy dispersive X-ray spectroscopy (EDX, Genesis XM2, EDAX, America). Phase composition of the Ni–B coatings was analyzed by X-ray diffraction (XRD, D8 Advanced, Bruker, Germany) with Cu Kα radiation (λ = 0.154 nm).

3. Results and discussion

Fig. 2(a) shows the morphology of original graphite powders. It can be seen that these powders show a bulk structure and consist of multiwall flake graphite. As shown in Fig. 2(b), graphite powders made a transition from bulk structure to flake structure after the pretreatment. Both original and pretreated graphite powders were uncovered. Because bulk graphite powders consist of multiwall flake graphite via weak van der Waals force, graphite flakes were easily torn off from bulk graphite surface by ultrasonic vibration during the pretreatment. This generated a better dispersion and larger free surface area of graphite. Consequently, the pretreatment effect was better. Wettability of graphite is important and was evaluated. A compact

Table 1
Chemical composition of electroless nickel plating bath.

Chemical	Formula	Concentration(g/L)
Nickel dichloride	$NiCl_2$•$6H_2O$	20
Citric acid trisodium salt dihydrate	$C_6H_5Na_3O_7$•$2H_2O$	80
Sodium borohydride	$NaBH_4$	6
Sodium hydroxide	NaOH	Appropriate amount

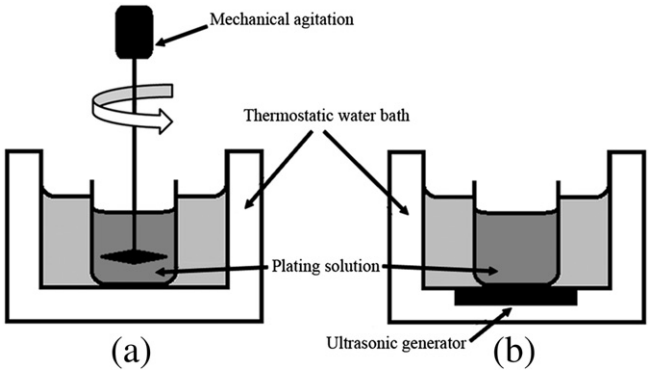

Fig. 1. Schematic diagram of experimental equipment of (a) mechanical agitation and (b) ultrasonic agitation.

graphite film with a thickness of mm scale was formed on ceramic plate by reciprocates and the contact angle was measured when distilled water-drop is standing on it. Fig. 3 shows the side-view optical images of water-drop on (a) original graphite film and (b) pretreated graphite film. The average contact angle between original graphite and water was 139 °C. However, the average contact angle between pretreated graphite and water was 56 °C. This shows that graphite made a transition from hydrophobicity to hydrophilic property after the pretreatment. The improvement of wettability contributes to the electroless plating on graphite.

After the pretreatment, graphite powders were introduced into nickel bath. In order to improve the properties of Ni–B coating, mechanical or ultrasonic agitation was used during the electroless plating. Both mechanical and ultrasonic agitations continuously provided fresh electrolytes to the graphite surface and prevented graphite from

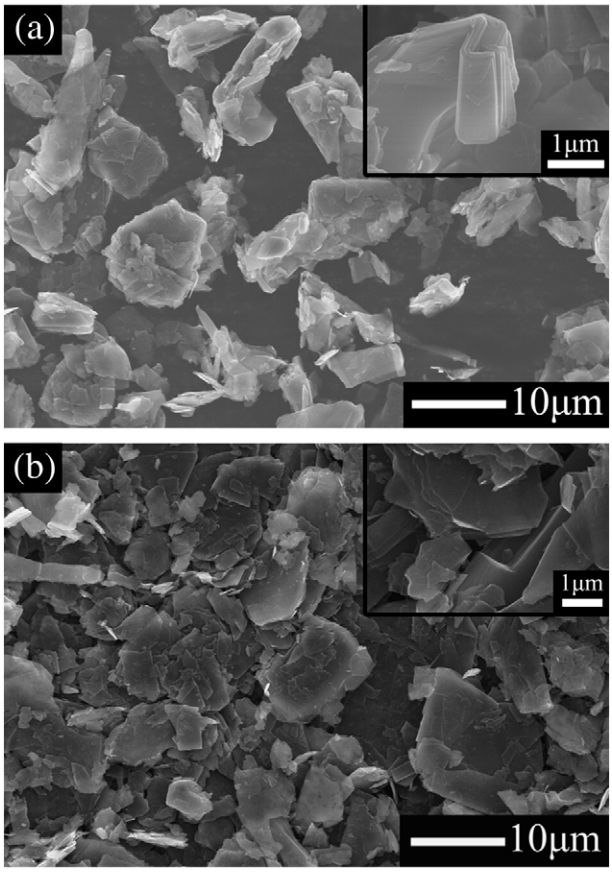

Fig. 2. SEM morphologies of (a) original graphite and (b) pretreated graphite.