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Preparation of a graphene–silver nanowire hybrid/silicone rubber composite for thermal interface materials

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ABSTRACT

In this study, we used acrylamide-modified reduced graphene oxide (AA-RGO) and thiophenol modified silver nanowires (mAgNWs) as reinforcements, and silicone rubber (SR) as a polymer matrix. One-dimensional mAgNWs and two-dimensional RGO can form synergetic effects and construct a three-dimensional network structure for thermal conduction in SR to improve the thermal conductivity of SR composites. SEM images showed a few aggregations of AA-RGO at 6 and 10 phr. However, the thermal conductivity of composites with some aggregations can be significantly higher than that of dispersed homogeneous composites. An interconnected hybrid network between AA-RGO and mAgNWs appears to have been caused by a π – π interaction. The thermal conductivity of mAgNW/AA-RGO/SR nanocomposites (approximately 1.152 W/mK) were higher than those of AA-RGO/SR composites without mAgNWs (approximately 0.412 W/mK), and considerably higher than those of neat SR (0.180 W/mK). Furthermore, the coefficient of thermal expansion (CTE) of the mAgNW/AA-RGO/SR nanocomposites were 148.3 ppm/°C, which differs considerably from the CTE of neat SR, 310.0 ppm/°C. Therefore, AA-RGO–mAgNWs hybrid/SR composites are promising carbon materials for thermal interface materials.

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1. Introduction

In past decades, consumer electronics have become indispensable to human life. To deliver lightweight highly efficient consumer electronics, integrated circuits (ICs) and central processing units (CPUs) with high levels of dissipated power are necessary. However, integrated circuits generate large amounts of heat [1]; thus, heat management is crucial. A typical method of dissipation is heat transfer by conduction to a heat sink. Heat exchangers are connected to heat sources in many industrial processes [2]. However, a device with a rough interface cannot attach completely and may only form multiple heat transfer paths. To solve this problem, thermal interface materials (TIMs) have been proposed to fill the air gaps of interfaces between devices [3]. Several types of TIM, such as greases, phase change materials, and filled polymer matrices, have been used in various industries [4].

The backbone of silicone rubber (SR) comprises repeating silicon to oxygen bonds; the silicon atoms at the ends of the backbone are also bonded to organic groups. SR differs from organic rubber, which possesses a carbon to carbon backbone, and SR can resist ozone, ultraviolet light, and heat in various extreme environments. SR is particularly known for its unusual properties; it is soft, stable, nontoxic, and nonflammable [5]. Therefore, it is widely used in various industrial applications, including automotive applications, electronics packaging, mechanical components, and biomedical devices [6]. However, typical SR has poor thermal conductivity, and is unsuitable for applications such as in TIMs. To improve the thermal conductivity of SR, high conductivity reinforcements can be incorporated into SR composites.

Graphene nanosheets (GNSs) are a two-dimensional hexagonal honeycomb carbon lattice forming a monolayer with a thickness of 0.34 nm. GNS possesses high specific surface area, good chemical modification, high electrical conductivity, and especially extremely high thermal conductivity of approximately 2300 W/mK [7,8]. GNS has attracted considerable attention because of its excellent properties and potential applications in many fields. Silver nanowires (AgNWs) have attracted extensive interest in

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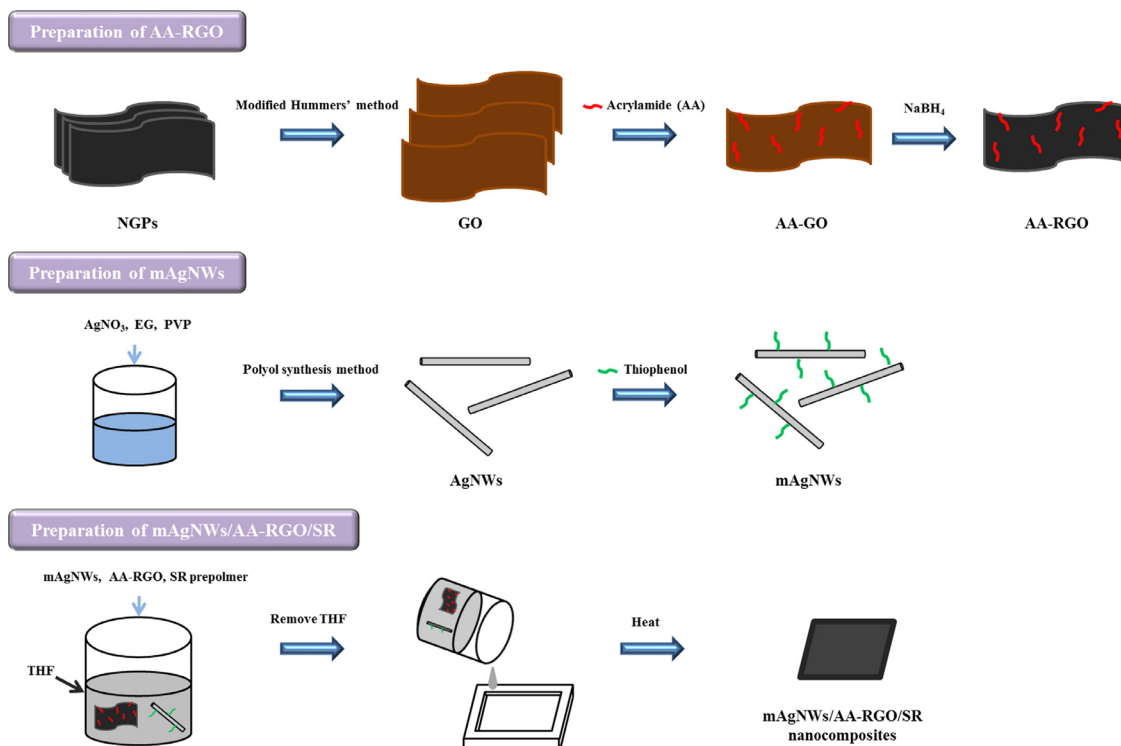


Fig. 1. Scheme of the procedure for preparing the mAgNW/AA-RGO/SR nanocomposites.

recent years because they have a nanoscale diameter, high aspect ratio, excellent thermal conductivity, and one-dimensional structure [9].

Hu et al. [10] indicated that incorporating multi-walled carbon nanotubes (MWCNT) and graphene into SR increases its thermal conductivity, because the synergic effect of the hybrid filler can improve the heat dispersion of SR. Pradhan and Srivastava [11] indicated that a combination of MWCNT and graphene in SR can be used for preparing nanocomposites with favorable mechanical properties and thermal conductivity. Goyal and Balandin [12] indicated that graphene and silver particles dispersed in epoxy resins can be used to manufacture TIMs. Previous studies have indicated that hybrid graphene/silver particles/epoxy composites have been observed to produce a significant enhancement of thermal conductivity.

With high GNS loading (20 wt%) in composites, restacking and irreversible agglomerations result from the π - π interactions among GNSs [13,14]. Agglomeration can increase the thickness of GNSs, but that tends to significantly decrease the thermal conductivity [15]. Therefore, this study proposed a method for developing hybrid fillers composed of one-dimensional AgNWs and two-dimensional GNSs that exhibit synergetic effects; this can be exploited to construct three-dimensional heat conductive networks and prevent the aggregation of GNSs in SR. When hybrid fillers with high thermal conductivity are added, SR nanocomposites can enhance their thermal conductivity beyond that of pure SR or SR nanocomposites prepared with individual fillers.

In this study, to enhance the interfacial compatibility between GNSs and SR, functionalized GNSs were prepared using acrylamide. To enhance the interfacial interaction between AgNWs and GNSs and to prevent the aggregation of GNSs, thiophenol was grafted onto AgNWs to form modified AgNWs (mAgNWs). The thiophenol on the surfaces of the mAgNWs possesses numerous phenyl functional groups, and was attracted by GNS because of the π - π interaction. This simple self-assembly process can be

employed to prepare AgNWs/GNS/SR nanocomposites with high thermal conductivity.

2. Experiment

2.1. Materials

Nanographite platelets (NGPs) produced through a chemical vapor deposition (CVD) process were provided by Angstrom Materials LLC (McCook Avenue Dayton, Ohio, US). The thicknesses of these NGPs were less than 100 nm. Sulfuric acid (H_2SO_4), potassium permanganate (KMnO_4), sodium nitrate (NaNO_3), hydrogen peroxide (H_2O_2), and ethylene glycol (EG) were received from the Showa Chemical Co. (Tokyo, Japan). Acrylamide (AA), N-(3-dimethylaminopropyl)-N-ethyl-carbodiimide hydrochloride (EDC), and tetrahydrofuran (THF) were obtained from Acros Organics Co. (Geel, Belgium). The chemicals for preparing the AgNWs were silver nitrate (AgNO_3) and polyvinyl pyrrolidone (PVP), which were received from Showa Chemical Co. Ltd. (Tokyo, Japan) and Acros Organics Co. (Geel, Belgium), respectively. Silicon rubbers (SR) were received from the Dow Chemical Company and Corning Incorporated (USA).

2.2. Preparation of GO

Graphene oxide (GO) was obtained by adopting a modified Hummers' method [16]. NGPs (0.5 g), NaNO_3 (0.4 g), and H_2SO_4 (25 ml) were fed into a three-necked 250-ml flask. After this was stirred in an ice water bath for 1 h at 0 °C, 2.25 g of KMnO_4 was added to the mixture slowly. Subsequently, the mixture was stirred at 50 °C until it turned to a highly viscous brown fluid. Next, 100 ml of RO water was added slowly at 80 °C, and 20 ml H_2O_2 was then added when the reaction was completed. The mixture was stirred continually for 2 h. To obtain a clean GO

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