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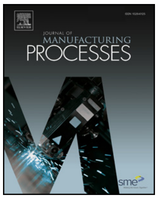

Influence of process parameters and temperature on the solid state fabrication of multilayered graphene-aluminium surface nanocomposites

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ABSTRACT

Electrical resistance heat assisted mechanical impregnation of nanomaterials can be effectively utilized as a solid state surface modification technique for soft materials like Aluminium. The quantity of heat generated at the interface and the extent of diffusion of nanomaterials depends on the contact pressure, electrode geometry, magnitude of current and the duration for which the current is applied. In this paper, we evaluate the influence of these process parameters on the mechanical impregnation of graphene on Aluminium substrates. The results are used to explain the experimentally observed microstructural and surface mechanical properties. The measured microhardness variation is in accordance with the temperature at various processing conditions and maximum surface hardness is observed at those conditions where the surface temperature is near the melting point of Aluminium. Influence of temperature on the possible strengthening mechanisms prevailing in Aluminium-graphene composites are elucidated in detail. Raman spectroscopic analysis provides insights into the temperature dependence of the change in intensities and shift in peak positions of respective G, D and 2D bands. X-ray diffraction analysis was carried out and the variation in lattice strain and crystallite size is correlated with the temperature data. The analysis presented herein provides encouraging results for realizing Aluminium-Graphene surface nanocomposites through a highly versatile, rapid and easily automatable solid state processing technique.

1. Introduction

In the past few decades, research in material science has become focused on composite materials as they can be developed into light weight, environment friendly, and high performance appliances. Aluminum and its alloys have been widely used for such applications, due to its excellent physical and mechanical properties, lightness, good electrical and thermal conductivity, corrosion resistance, suitability of surface treatments, and ability to recycle. However, Aluminium surface is poor in certain other mechanical properties like low tensile strength, low abrasion resistance, susceptible to corrosion and softness [1,2]. In this scenario, production of aluminum alloys and composites with a decreased specific density and improved mechanical and metallurgical properties is one of the most significant goals of contemporary material science investigations. Carbon nanomaterials, particularly graphene, have been extensively used as reinforcement of aluminum, to meet those high and ever increasing demands [1–4].

The typical methods used to fabricate Aluminium-Graphene Metal matrix composites include Powder Metallurgy [5,6], Semi-Powder Metallurgy Method [7] and Squeeze Casting Method [8]. In powder metallurgy route, commercial colloidal Graphene Oxide is coated onto Aluminium powder particles and then reduced via thermal annealing to get reduced Graphene [6]. Semi-powder metallurgy method is actually a powder metallurgy method followed by hot extrusion technique. Hot extrusion is done to have more even mixing and stronger reinforcement [7]. In squeeze casting method, formation of a preform and its infiltration with Aluminium alloy is involved. Proper portions of components are mixed in aqueous solution of the inorganic binder. The homogeneous mixture is drained off and formed to desired shape [8]. Another method of making Aluminium-Graphene composite was reported using different base powder preparation in powder metallurgy [9]. Graphene oxide (GO) sheets with the negative charge were prepared by a modified Hummers' method and Al powders were coated by hexadecyl trimethyl ammonium bromide (CTAB) to obtain the surface positive charge. Then, GO–Al powders are obtained by electrostatic self-assembly to realize the homogeneous adsorption of GO sheets on Al powders. Finally, Graphene reinforced aluminum matrix composites were fabricated by powder metallurgy [9].

The Aluminium- Graphene composite can also be obtained by direct chemical interaction of carbon-containing substances “in situ” with

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molten aluminum under a layer of molten alkali halides in air [10]. The synthesis of Graphene nanosheets in a metal matrix is one-step, simultaneous process, taking place directly in molten aluminum under alkali halides melt without the necessity of a separate stage of synthesis and introduction of Graphene. The Aluminum-Graphene composites formed according to this method are characterized by a high uniformity of Graphene films [10]. The primary aim of all the processes is to combine the superior features of stiffness and conduction of the Graphene with Aluminium. However, all the reported works were focused on fabricating bulk composites at processing temperatures well above the melting point of Al. Not many studies, other than friction based techniques [11,12], were reported on fabricating Al-Graphene surface nanocomposites where the surface properties are modified keeping the bulk properties of Al intact.

Electrical Resistance heat assisted mechanical impregnation is a novel solid state processing technique to cause diffusion of Graphene and other nanomaterials onto Aluminium and it successfully circumvents these problems [13,14]. In this method, mechanical pressure is applied on graphene coated Aluminium substrates which are locally softened by Joule's heating. The heat generated at the Graphene-Aluminium interface will act as the driving force for the diffusion process. The interface temperature is maintained higher than recrystallization temperature of aluminum but lower than its melting temperature. The number of atoms diffusing increases exponentially with temperature as given by the Arrhenius relation. Experimental results have shown that Graphene coated surfaces are characterized by a higher mechanical properties, abrasion resistance, and corrosion resistance as compared to unmodified Aluminium [13–15]. In this context, the objective of the present work is to do a detailed thermal analysis to evaluate the influence of temperature and processing conditions on the microstructure and surface mechanical properties. Experiments are conducted at various processing conditions (current & time) and the microstructure and surface mechanical properties are evaluated. A finite difference model has been developed and mathematical simulation using MATLAB has also been carried out. The temperature distribution at affected areas for different values of current, electrode force, electrode tip diameter and time are evaluated and correlated with the surface hardness data.

2. Materials and methods

2.1. Materials

For the experiments Aluminium-1100, which contains approximately 98.2% Al, 0.75% Si, 0.83% Fe and minor quantities of other elements, was used. The plates of thickness 1 mm were cut into 100 × 100 mm size and coated with graphene nano-platelets (GNP) dispersed in Polyvinyl alcohol (PVA) solution by drop casting. The thickness of the so obtained graphene coating was approximately 100 μm. The Graphene flakes, which is a mixture of single layer (A), multilayer (B) and aggregates (C) (Fig. 1), was purchased from United Nanotech Innovation Pvt. Ltd. with an average dimension in X and Y direction: 10–20 micron, average thickness: 3–6 nm, average number of layers: 6–10, tensile modulus parallel to the surface: > 1000 GPa and tensile strength: > 5 GPa. Polyvinyl alcohol (PVA) was purchased from LOBA Chemie with the following properties. Degree of polymerisation: 1700–1800, Viscosity: 25–32cps and pH (0.2% in water): 5–7.

2.2. Experimental setup for solid state processing

For the electrical resistance heating and pressing, a spot welding machine supplied by ELECTROWELD (Model No: SP30PR) was utilized [14,16,17]. The machine operates at a main voltage supply of 415 V, with maximum available short circuit current 12 kA and maximum power output (at 50% duty cycle) of 30 kVA. The setup consists of two copper electrodes between which the sample is kept and current is passed, as shown in Fig. 2. The electrode serves multiple purposes like carrying current, holding the sample and applying force. The setup produces a thermo-mechanical effect, which is utilized here for the mechanical impregnation and diffusion of graphene. The force applied on the electrode, which is regulated by means of air pressure, is adjustable and gives a constant force from 1 to 4 K g/cm^2. To impregnate the graphene coated over the substrate, the coated aluminium sample is placed between the copper electrodes. The force acting on the sample surface by the electrode is kept constant for a particular experiment. A fixed constant current (1000/ 2200 /3000 A) is applied for a small time duration (0.1/ 0.3/ 0.5/ 0.7 s). The amount of heat generated at any point on the cross section will depend on the current passed, electrical resistance offered and the time for which the current is applied. If the current and time conditions are kept constant then the point which offers maximum electrical resistance will be affected by the maximum heat input [18]. After the impregnation process, the sample is taken out and kept aside for cooling at room temperature. The surface is further cleaned with distilled water and acetone to dissolve the PVA and to remove the un-impregnated graphene at the surface. The impregnation depth varies in the range 50–100 μm, depends on process parameters.

2.3. Characterization

The surface profile of GNP coated Al sample was analysed on a 3D-Optical Scanning Profilometer (OSP) manufactured by Bruker (Model: Contour GT, Max scan speed: 47 μm/sec, RMS repeatability: 0.02 nm, lateral resolution: 0.38 μm min (sparrow criterion) and 0.26 μm (with Acuity XR), step height accuracy: <0.75% and step height resolution: <0.1%). The average surface roughness (R$_a$) was found to be 4.433 μm. Fig. 3 shows the surface profile of GNP coated Al obtained from OSP. Micro hardness test was carried out to determine the hardness values (Hv) of graphene impregnated aluminium processed at various conditions using OMNITECH-S-AUTO. The indentation load was 200 gm and a dwell period of 10 s was provided. All the measurements were carried out at room temperature. Field emission scanning electron microscopic (FE-SEM) analysis was carried out on ZEISS MERLIN instrument at an operating voltage of 5 kV. JEOL model JEM2100 which has a lattice resolution of 0.14 nm and a working voltage of 200 kV was used for the transmission electron microscopy (TEM) analysis. X-Ray Diffraction (XRD) analysis of the processed samples was done using PANalytical XPERT PRO with CuKα with 2θ range 5°–85°. X-pert Highscore software was used for the detailed analysis of the obtained XRD data. The crystallite size (a) was calculated using Scherrer's equation, $a = K\lambda/\beta Cos(\theta)$, where, K is the Scherrer's constant ($K = 0.9$), λ is the wavelength of the incident X-ray, β is the full width at half maximum (FWHM) of individual peak after deducting the instrumental broadening and θ is the Bragg's diffraction angle. For Raman spectroscopic analysis, HORIBAJOBINYVON T6400 Raman Spectrometer with Olympus microscope was used along with a 514 nm ArCr laser.

3. Results and discussion

3.1. Heat flux and temperature distribution

Heat flux and temperature values in this study are not experimentally determined but are calculated by using numerical methods. An axisymmetric finite element model was developed to predict the temperature distribution and the details of the model are given in the supplementary information. The heat flux/temperature simulation is done only for bare aluminum and this simulated value is used as a "measure" of the temperatures encountered in an Al substrate impregnated with a thin layer of graphene. This assumption is valid since the graphene layer is thin and only comprises of ∼0.1–0.3% of the volume.

Variation in heat flux with respect to the input current at various values of contact force F and electrode diameter d is summarized in Fig. 4. (The governing equations for calculating the heat flux and the