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A novel in-situ polymer derived nano ceramic MMC by friction stir processing

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ABSTRACT

Friction stir processing (FSP) is a solid state technique used for material processing. Tool wear and the agglomeration of ceramic particles have been serious issues in FSP of metal matrix composites. In the present study, FSP has been employed to disperse the nanoscale particles of a polymer-derived silicon carbonitride (SiCN) ceramic phase into copper by an in-situ process. SiCN cross linked polymer particles were incorporated using multi-pass FSP into pure copper to form bulk particulate metal matrix composites. The polymer was then converted into ceramic through an in-situ pyrolysis process and dispersed by FSP. Multi-pass processing was carried out to remove porosity from the samples and also for the uniform dispersion of polymer derived ceramic particles. Microstructural observations were carried out using Field Emission Scanning Electron Microscopy (FE-SEM) and Transmission Electron Microscopy (TEM) of the composite. The results indicate a uniform distribution of ~100 nm size particles of the ceramic phase in the copper matrix after FSP. The nanocomposite exhibits a five fold increase in microhardness (260HV$_{100}$) which is attributed to the nano scale dispersion of ceramic particles. A mechanism has been proposed for the fracturing of PDC particles during multi-pass FSP.

1. Introduction

Metal matrix composites (MMCs) combine the ductility and toughness of metal with the high strength and elastic modulus of ceramic. MMCs have potential applications in automotive, aeronautical and aerospace industries [1,2]. Hence, simple and economical methods for fabricating MMCs are the most sought after research area [3]. In MMCs, damage evolution starts preferentially at particle clusters in the matrix [4]. This is due to the higher local particle volume content and the higher stress triaxiality [5]. Problems such as fatigue fracture and impact resistance, ductility, agglomeration along with the predictability of properties of MMCs have been the major issues that have limited the use of MMCs. In order to overcome these shortcomings, the use of nano particles has been attracting increasing attention in composite research. The reason is their capability in improving the mechanical and physical properties of traditional MMCs. The dispersion of a *nanoscale* ceramic phase is needed in order to overcome the problems related to fatigue, fracture toughness, and creep behavior at high temperatures [6]. However, manufacturing costs, preparation of nano composites and environmental concerns have to be addressed.

MMCs/nano MMCs have been produced through the powder metallurgy route [7]. Secondary processing, such as extrusion and rolling are often needed to fully consolidate materials produced in this manner. A high extrusion ratio is often required to disrupt the oxide film between metal and powder particles. An alternative route is to introduce ceramic particles by stirring them into molten metal [8]. However, the nanoscale particles form hard agglomerates that are difficult to disperse through physical mixing [9].

Recently, a new method of introducing polymer derived ceramics (PDC) into metals from the polymer route has been developed. The polymer precursor is dispersed in the metal and then converted in-situ to a ceramic phase. The polymer can be introduced through casting or the powder metallurgy route. In the casting route, the polymer powder is directly added to molten metal and pyrolyzed in-situ to create castings of metal-matrix composites. These composites have shown better properties at elevated temperatures but the problem of agglomeration and porosity still remains [10]. In the powder method, the organic precursor was milled with copper powder and then pyrolyzed to produce a metal matrix composite. This retains its mechanical strength close to the melting point of the copper matrix and this is because the ceramic phase does not coarsen. But, getting a nano sized distribution is difficult through this route [11].

Friction stir processing (FSP) has successfully evolved as an alternative technique to fabricating metal matrix composites [12]. FSP is based on the principles of Friction Stir Welding (FSW) [13]. In FSW, a rotating tool with a pin and a shoulder is inserted into the material to be joined, and traversed along the line of the joint. The friction between the tool and the work piece result in localized heating that softens and plasticizes the material. The role of the friction stir welding tool on the material flow

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has been studied in detail by Kumar et al. [14,15]. In the FSP of MMCs, the material undergoes intense plastic deformation resulting in the mixing of ceramic particles and the metal. FSP also results in significant grain refinement [16] and has also been used to homogenize the microstructure of nanocomposite aluminum alloys [17]. FSP technology has also been used to refine the microstructure of cast aluminum alloys, iron and particle reinforced composites [18,19], fabricate a surface/bulk composite of Al–SiC on an aluminum substrate [20–22], friction stir surfacing of cast aluminum silicon alloy with boron carbide and molybdenum disulfide powders [23] and produce ultra-fine grained Cu/SiC composites [24].

The major problem in the FSP of MMCs is the severe tool wear which results from abrasion with hard ceramic particles. The progressive wear of the tool increases the likelihood of void development [25,26]. Excessive wear can alter the tool geometry as reported in the friction stir welding of aluminum alloy 6061 containing alumina (Al_2O_3) particles [27]. The problems concerning the tool life, especially when abrasive particles are involved [26], have become a serious issue in the application of FSP as a development of MMCs for applications in aerospace, automotive and marine systems.

In the current study, a new method has been developed by authors to produce polymer derived nano ceramic MMCs through FSP. This methodology has proved successful in addressing the aforementioned issues in fabricating MMCs. This method mainly consists of three steps. In the first step, the mixing of a polymer with metal is carried out by FSP. In the second step, in-situ or external pyrolysis (as per requirement) of the polymer reinforced FSP MMCs and finally another pass of FSP on the same for removing porosity and uniform dispersion of polymer derived ceramic particles in the matrix are carried out and there produce nano-scale metal matrix composites with a high volume fraction of the ceramic phase. This method can easily mix the polymer, pyrolise the polymer, remove the porosity, refine the ceramic particles and make nano-size dispersion in MMC. This method can be extended to big sheets or a particular region in a sheet with no or low wear of tools. The polymer, being malleable, breaks down into fine particles which are then converted into a ceramic phase by an in-situ pyrolysis at 800 °C. The ceramic phase is redistributed again by further processing. In the present experiments, we seek to introduce approximately 20 vol.% of the ceramic phase into copper. The microstructure and mechanical properties of the developed copper-based in-situ polymer derived nano MMCs have been characterized in detail to understand the distribution of particles and a method has been proposed for the fracture of PDC during FSP.

2. Experimental procedure

The material selected in the present study was pure copper (99.9%). The polymer precursor was poly (urea methyl vinyl) silazane, which is available commercially as CERASET (CERASET is a trademark of Kion Corporation, Charlotte, NC.). It consists of silicon, carbon, nitrogen, oxygen and hydrogen. The liquid precursor was thermally cross-linked into a rigid polymer, which was milled into a powder. This powder, having angular shaped particles of an average size of 10 μm was measured by Mastersizer S Ver. 2.18 and was used as reinforcement. The powder is a cross-linked polymer having a density of ~1.0 g cm^{-3}.

Grooves (3 mm × 4 mm) were cut and holes (ϕ3 mm) were drilled into a 6 mm thick copper plate and were filled with the polymer powder. For multi-pass FSP, the tool pin was moved along one single line, and successive passes were applied after the work piece had been cooled down from the previous FSP pass. The experiments were carried out in ambient conditions as the powder quickly incorporated within the copper which prevents oxidation. The detailed procedure is also described in [28].

The FSP experiments were carried out in a five axis friction stir welding machine (BiSS — ITW, Bangalore). A densimet tool with a frustum shaped threaded pin, 6 mm top diameter, 4 mm bottom diameter, rounded end and 25 mm diameter flat shoulder with a chamfered edge was used. The tool was tilted at 3°. The tool was rotated in a counter clockwise direction at 1500 rpm and traversed with a speed of 25 mm/min.

A differential scanning calorimetry (DSC) in TGA mode (Perkin-Elmer DSC-7) was carried out in a temperature range of 10–1000 °C at a

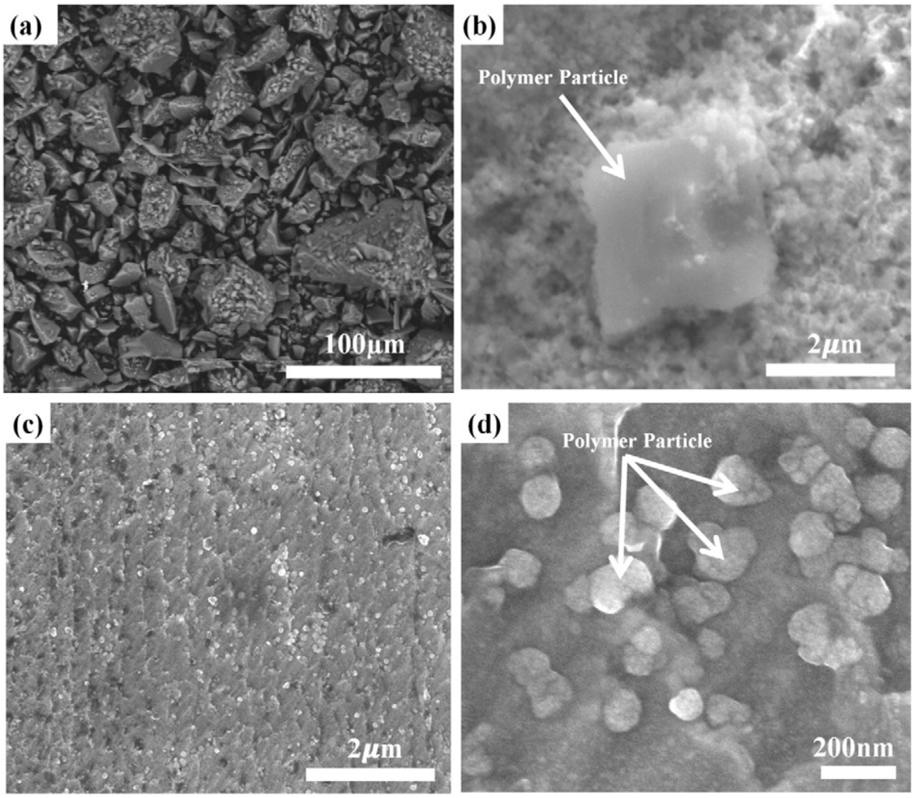

Fig. 1. SEM image of (a) As received SiCN polymer powder, (b) first pass FSP Cu/PDC shows SiCN polymer particle, (c) four pass FSP Cu/PDC shows uniformly dispersed nano polymer particles and (d) high magnification image of nano polymer particles after four passes [Images were taken at 5–10 kV].