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Exceptional micromachining performance of silicon carbide ceramics by adding graphene nanoplatelets

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ABSTRACT

The electrical discharge machining (EDM) performance of silicon carbide (SiC) ceramics containing graphene nanoplatelets (GNPs) is investigated for the first time. Under fine machining conditions, the material removal rate (MRR) dramatically increases up to 186% when 20 vol.% of GNPs are added to SiC ceramics, leading to reductions on the electrode wear rate of 132%. The EDMed nanocomposites exhibit surface roughness $\leq 0.8\,\mu m$. This outstanding EDM response of the graphene nanocomposites as compared to monolithic SiC is explained by their enhanced transport properties, establishing a direct dependence of MRR with the electrical conductivity. EDM performance of the nanocomposites also depends on the testing direction for materials with low GNPs connectivity (≤ 10 vol.%). Melting/evaporation are the main removal mechanisms, thermal spalling also operating for low thermal conducting materials. The employ of EDM on SiC/graphene nanocomposites allows machining microparts with a fine dimensional precision, opening new opportunities for SiC-based microcomponents.

1. Introduction

The development of silicon carbide (SiC) ceramic microcomponents to be used, among others, as part of microturbines, microreactors, and microelectromechanical systems or as catalytic microsupports is attracting a great interest mainly due to the excellent thermal, tribological and mechanical performances of these ceramics, jointly with their good resistance to corrosive and harsh environments [1,2]. However, the machining of SiC complex microparts is a complicated task when using diamond grinding wheels, the most common technique, due to the high hardness and brittle nature of SiC, which leads to expensive and time consuming processes for getting microcomponents with relatively low accuracy and surface finishing.

Electrical discharge machining (EDM) arises as one of the most suitable methods to overcome these difficulties, since the material removal is caused by electrical discharges, and mechanical forces between the electrode and the workpiece are not developed [3].The main constraint for EDM is that a minimum electrical conductivity (σ_e) of the workpiece is required ($> 0.3–1.0\,S\,m^{-1}$) to enable the electrical discharges [4], which can be a clear limitation in the machining process of SiC ceramics typically exhibiting lower conductivity values than those needed. Ferraris et al. [5] and Pachaury et al. [6] have recently reviewed the different approaches to enhance the discharge efficiency of ceramics, including SiC ones. In this way, Fukuzawa et al. [7] proposed the use of an assisting electrode method (AEM) to machine insulator ceramics by coating the workpiece with a conductive layer. This layer promotes the first discharges between the electrode and the workpiece. The pyrolytic carbon generated during the decomposition of the oil-based dielectric fluid adheres to the ceramic surface, leading to the continuous formation of an intrinsic conductive layer on the workpiece that allows a successful EDM process. Although AEM has been employed for non-conductive SiC ceramics [8,9], the material removal rate (MRR) was low ($\sim 10^{-3}\,mm^3\,min^{-1}$) [9] and the electrode wear rate was quite high. Another approach to promote the EDM of low electrical conductive materials consisted in the addition of conductive powders into the dielectric fluid, known as powder mixed EDM (PMEDM) process [10]. In particular, Liew et al. [11] improved the electrical discharge frequency and the EDM performance of reaction bonded-SiC (RB-SiC) ceramics ($\sigma_e = 6.9 \times 10^{-2}\,S\,m^{-1}$) by incorporating electrical conductor carbon nanofibers to the dielectric fluid. The best surface quality of the machined part ($R_a \sim 0.2\,\mu m$) was attained with nanofibers concentration of around $0.02\,g\,L^{-1}$, getting low MRR values, in the order of $10^{-4}\,mm^3\,min^{-1}$. Finally, successful EDM attempts were car-

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ried out increasing the electrical conductivity of SiC materials. For instance, Clijsters et al. [12] machined complex RB-SiC parts using the die-sinking EDM technology thanks to the high σ_e ($\sim 10\ S\ m^{-1}$), which was due to the silicon remaining within the ceramics after the infiltration process. These authors obtained high MRR ($3.6\ mm^3\ min^{-1}$) with a R_a value of 2.9 μm when used roughing settings; whereas MRR considerably decreased to $10^{-2}\ mm^3\ min^{-1}$ with $R_a = 1.1$ μm for tests carried out under finishing parameters. Other strategies to turn SiC ceramics into conductive materials consisted in N-doping the SiC lattice by introducing yttrium nitrate as sintering additive [13]. Using this approach, Yoo et al. [13] achieved σ_e values of $\sim 10^4\ S\ m^{-1}$ that allowed the EDM of quite complex SiC shapes with a MRR of $\sim 2 \times 10^{-3}\ mm^3\ min^{-1}$.

Following this last strategy, graphene, in the form of graphene nanoplatelets (GNPs) or graphene oxide (GO) sheets, has proved to be an extraordinary filler for enhancing the electrical response of low conductor ceramics such as Al_2O_3 [14], ZrO_2 [15], Si_3N_4 [16] or B_4C [17]. Therefore, ceramic/graphene composites are promising materials to be efficiently EDMed. At present, few EDM studies with limited data, which do not allow a sound comparison among them, have been carried out for Si_3N_4 [18], B_4C [17] and Al_2O_3 [19] ceramics containing GNPs fillers. In this way, Hanaoka et al. [18] successfully EDMed high electrically conductive Si_3N_4/20.6 vol.% GNPs nanocomposites, reporting a decrease of the electrode wear rate (~84%) and the surface roughness (~50%) of the machined parts as compared to the reference Si_3N_4 ceramics, an insulator material that required the employ of AEM. Tan et al. [17] considerably enhanced the machinability of B_4C by incorporating up to 5 vol.% of GNPs, just reporting increases in the MRR of up to ~600%. Finally, Sung et al. [19] simply found a substantial decrease on the surface roughness of micro-EDMed Al_2O_3/GNPs nanocomposites as the GNPs content increased from 10 to 20 vol.%. Therefore, a deeper analysis on the EDM performance of ceramic/graphene nanocomposites is needed.

As some of the present authors have demonstrated, the electrical conductivity of SiC ceramics can be also increased in three orders of magnitude by introducing GNPs [20], reaching maximum σ_e values of $\sim 4 \times 10^3\ S\ m^{-1}$. Besides, graphene fillers also provided multifunctionality to SiC, enhancing both their tribological [21] and mechanical [22] responses. However, to the best of our knowledge, EDM has not been employed to manufacture SiC/graphene parts hitherto.

The goal of this work is, hence, to investigate the EDM performance of SiC/GNPs nanocomposites with distinct GNPs contents (10 and 20 vol.%) comparing their responses with three different monolith SiC materials, two of them containing graphene flakes in-situ grown during the sintering process of the ceramics. The selection of this set of materials scanned a wide range of properties, especially in terms of the electrical and thermal conductivities. This allows us establishing a relationship between the EDM response and the transport properties of the materials. In addition, the machining experiments were conducted varying the energy conditions, and the graphene-based nanocomposites were tested for the first time on orthogonal surfaces according to their anisotropic microstructures.

2. Experimental procedure

2.1. Materials fabrication

Five different SiC-based materials were chosen to explore their EDM performances (Table 1), in particular, three monolithic SiC ceramics showing distinct properties and two SiC/GNPs nanocomposites containing 10 and 20 vol.% of GNPs. Specimens of one of the monolithic SiC ceramics were commercially manufactured (CD110 grade, CeramTec, Germany). The rest of compositions fully dense specimens were produced in-house according to the experimental procedure described elsewhere [20,23]. In brief, SiC/GNPs powders were processed by mixing and sonicating two isopropyl alcohol suspensions independently prepared: one containing commercial GNPs (type N006, Angstron Materials Inc., USA) that were dispersed through a sonication process; and another SiC-based suspension consisting on the attrition milled ceramic powder composition −93 wt.% of micro-sized β-SiC (BF-17A, H.C. Starck, Germany), 5 wt.% of Y_2O_3(Grade C,H.C. Starck, Germany), and 2 wt.% of Al_2O_3 (SM8, Baikowski Chimie, France). The dried and sieved SiC/GNPs powder mixtures (labelled as 10GNPs and 20GNPs for 10 and 20 vol.% GNPs contents, respectively) were then spark plasma sintered (SPS, Dr. Sinter, SPS-510CE, Japan) into disc specimens of 20 mm diameter × 3 mm height at 1800 °C for 5 min, applying a uniaxial pressure of 50 MPa during the heating cycle, and using a vacuum atmosphere of ~6 Pa. Monolithic SiC specimens (0 vol.% GNPs) were equally processed from the ceramic powders using the above mentioned micro-sized β-SiC as well as a nano-sized β-SiC powder (NanoAmor, USA). Accordingly, the manufactured monolithic SiC ceramics were labelled as μ-SiC and *n*-SiC, respectively; meanwhile the commercial one was identified as C-SiC. Table 1 collects the different materials and their main properties.

It is important to remark that the SPSed μ-SiC and *n*-SiC ceramics contained ~3–4 vol.% of graphene multilayers, which were in-situ grown at the SiC grain boundaries during the sintering process [23]. Besides, both types of graphene fillers, the in-situ grown and the added GNPs, appeared into the material preferentially oriented with their basal (ab) plane perpendicular to the SPS pressing axis [20] (see an example in Fig. 1a), leading to materials with anisotropic properties (Table 1).

2.2. EDM tests

The EDM trials were performed using a SARIX micro-EDM machine (Model SX-200-HPM, Switzerland). A tungsten carbide rod with a diameter of 300 μm was used as tool electrode and microgrooves were machined into the materials with the rotating microrods. The tests were carried out using a machining depth and length of 50 μm and 200 μm, respectively, and an infeed of 10 μm. IME 110 (Oelheld GmbH, Germany) was used as dielectric fluid. In the case of *n*-SiC ceramics and 10GNPs and 20GNPs nanocomposites, their σ_e values (Table 1) were well above the limit ($\geq 1\ S\ m^{-1}$) for directly using the EDM process. However, for the low electrical conducting C-SiC and μ-SiC ceramics, it was not possible to employ EDM and AEM was required to enable the machining process. In this way, C-SiC and μ-SiC specimens were coated by screen printing with a conductive carbon lacquer layer of ~25 μm of thickness to act as assisting electrode, promoting the first sparks between the electrode and the carbon coated SiC workpiece. Those sparks would decompose the oil-based dielectric fluid, creating a continuous pyrolytic carbon layer on the workpiece that enables the machining process.

To explore the EDM performance of the ceramics and nanocomposites, three different EDM energy conditions were selected (Table 2) from preliminary trials that enabled a stable EDM process in all materials. In particular, the selected energy settings corresponded to conventional fine machining (EDM-F) and rough and fast machining (EDM-R1 and EDM-R2) processes. In the case of EDM-R1 and EDM-R2, the generator only allows one discharge per pulse width, while for EDM-F a larger number of discharges having shorter on-times takes place. The main difference between rough conditions is that the applied voltage is higher for EDM-R2 (Table 2). All the materials were EDMed in the (||) direction (Fig. 1b). In addition, EDM tests were also carried out in the (⊥) direction (Fig. 1b)

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