

# **A NOVEL PROCESSING TECHNIQUE FOR METAL-CERAMIC COMPOSITES**

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## A NOVEL PROCESSING TECHNIQUE FOR METAL- CERAMIC COMPOSITES

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### INTRODUCTION

Powder-based metal-ceramic composites can be produced by a variety of methods including the use of large electrical currents. For over fifty years it has been known that the direct electrical resistance heating of conductive metallic powder materials can cause densification. The mechanism by which this densification proceeds is material specific and is related to time at temperature during processing. The interest in the use of electrical current for heating of powders has been sporadic [1-5] and has been handicapped by the absence of a well-matched power source.

Over the last decade, devices for the production of pulsed electrical power have enjoyed a developmental thrust. In particular, homopolar generators (HPG) based upon the Faraday disk have been successfully engineered and commercialized[6]. The availability of these machines as pulsed power sources has fostered the development of novel powder processing approaches for metal-ceramic composites.

At the University of Texas at Austin, powder processing powered by a homopolar generator has been developed as a high-energy high-rate (HEHR) materials processing technique[7]. The '1 MJ in 1s' pulsed energy delivery from a 10 MJ HPG has been employed in a wide-ranging series of processing experiments. Metal-ceramic composite processing has been studied extensively [8-11]. The material systems characterized have included aluminum-silicon carbide[8], nickel-molybdenum boride[9], and copper-graphite[10, 11].

In what follows we will describe in detail the processing approach employed for the consolidation of copper-graphite composites, and the results of their evaluation as high temperature tribological materials.

In order to obtain a maximum electrical conductivity in a copper-graphite composite material, a continuous three dimensional copper network must be formed. Two manufacturing techniques for conventional copper-graphite composites are powder metallurgy and metal infiltration. In the conventional P/M technique, uniform mixing is difficult to achieve due to the large difference in density between the copper and graphite constituents. A thin graphite coating which develops on metal particles during the mixing prevents direct metal to metal contact, resulting in a low strength matrix. In the infiltration technique, the metal fillers may not be continuous in the porous graphite structure after cooling due to the differences in thermal contraction.

Due to the equilibrium immiscibility between copper and graphite at all temperatures, the interfacial bonds are weak; they can be improved most easily by adding low melting temperature metal binders, such as tin and lead. These increase the wetting between the matrix and graphite particles in the conventional sintering process. However, when used in high-current, high-speed sliding contact applications the flash temperatures at the sliding contact interface are so high that the low melting temperature constituents of the brush materials are extremely undesirable. In fact, the melting of the binder phase in a P/M copper-graphite brush was observed at the sliding speed of 220 m/s without carrying any current[12]. In addition, the electrical conductivity of copper is very sensitive to the impurities. The large interfacial electrical power loss associated with high current density and contact resistance can be partially offset by minimizing the brush electrical resistivity, as recently described in a contact resistance model[13].

For these reasons, binderless copper-graphite composites with sufficient strength are the most desirable candidates for high-current high-speed brush materials.

High-energy high-rate consolidation employing a homopolar generator is an attractive alternative processing approach for binderless copper-graphite composites based on the following characteristics: (1) extremely fast processing to minimize time for internal oxidation; (2) rapid heating and cooling rates associated with the pulsed Joule heating; (3) possible microencapsulation by preferential heating and local melting at copper-graphite interface; (4) a dense product with improved mechanical, electrical and thermal properties can be produced by simultaneous forging during the discharge.

## EXPERIMENTAL APPROACH

**Materials:** Studies indicate that for a binderless copper-graphite high-current brush material, the graphite content should not be less than 10 w/o, while the copper matrix mass fraction should be as high as possible. In this HEHR binderless copper-graphite composite brush consolidation, the graphite content is thus selected as 11 w/o. The copper and graphite powders used are the same as in a commercial P/M sintered composite which contains 5 w/o tin, 2 w/o lead as binders, and 11 w/o graphite. The appearance of the starting powders is shown in figure 1, which indicates the morphology and size distribution of the dendritic copper powders and the flaky graphite.

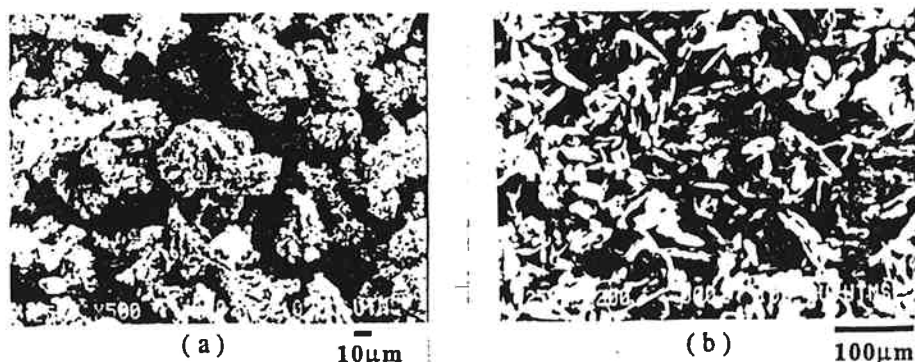


Figure 1 SEM micrographs of (a) dendritic copper powder and (b) flaky graphite powder.

**The Consolidation Process:** Figure 2 shows the schematic tooling configuration of the HEHR consolidation process. Investigations have involved the use of a 10 MJ pulsed HPG as a power source and a modified vertical axis hydraulic press with a 890 kN(100 ton force) capacity. The operating characteristics of the HPG are described elsewhere [14]. Powders are mechanically mixed and then loaded into the cavity of the die which was made of a dense alumina tube with an inner diameter of 32 mm. After compacting the powders to a desired pressure, the large current pulse was discharged from the HPG, passing through the powder mixture.

Following the discharge, the pressure was maintained for 180s. This holding period permits conductive heat transfer through the plungers to massive copper platens. The die was then unloaded and the consolidated compact was ejected. The current pulse length ranged from 1 to 3 seconds, depending on both the discharge rpm and the external system resistance. The peak current ranged from 31kA to 136 kA, depending again on the above two parameters. The external system resistance was mainly composed of the electrode (plunger) resistance and the resistance of the compact. The interfacial resistances are considered negligible within the pressure range applied (241-689 MPa).

Table 1  
THE PROCESS PARAMETERS EMPLOYED

COMPACTING PRESSURE:	241-689 MPa
HPG DISCHARGE RPM:	880-1300 (0.15-0.25 MJ)
DIE DIAMETER:	32 mm
POWDER MASS:	15g OR 30g
L/D RATIO:	0.1 OR 0.15
ELECTRODE:	ETP Cu OR 304SS

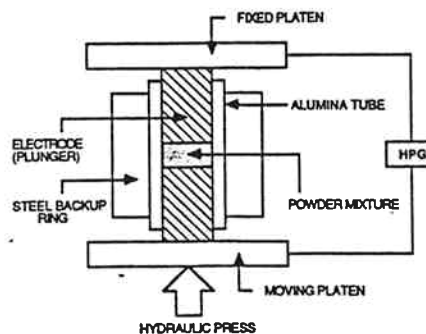


Figure 2 Schematic configuration of the HEHR consolidation process.

**Process Parameters:** In this series of experiments, material variables were kept fixed, i.e., composition (11 w/o graphite) and type of powders (proprietary copper and graphite powders). Emphasis was given to the effects of process parameters on the consolidated compact properties. The process parameters used are described in table 1.

## RESULTS AND DISCUSSION

**General Features of Consolidated Compacts:** Three general features were observed for HEHR consolidated compacts:

(1) **Preferred orientation** : Based on x-ray diffraction patterns and microstructure examination the basal planes of graphite platelets were found to be normal to the compacting direction, except for the limited region near the edge which had been back extruded. The uniaxial compression with large height-to-diameter (L/D) ratio is responsible for this phenomenon.

(2) Anisotropy (axial vs radial) : Electrical resistivity and hardness measurements showed anisotropy between axial and radial directions of the compact. This was mainly attributed to density variation due to stress and temperature distributions inside the die during the consolidation, which led to a greater degree of densification in the axial direction [3,15].

(3) Absence of oxidation : Although all processing was conducted in air, the short process time at high temperature minimizes the degree of internal oxidation. An as-processed pure copper compact which had a near theoretical density showed a 91% electrical conductivity relative to OFHC copper. Hydrogen annealing of this compact would further enhance the conductivity.

*Effect of Compacting Pressure:* In figure 3 a linear relationship between the final compact density and the compacting pressure is observed. As the pressure increases from 241 to 689 MPa, the compact density increases from 6.30 to 6.68 g/cm<sup>3</sup> which is 99.85% of the theoretical density. Figure 4 shows that the hardness which is the average of the hardnesses measured along the radial direction on the compact end surfaces also increases with the compacting pressure, as a result of density increase. It is obvious that the compacting pressure plays a key role in the copper-graphite composite consolidation when the discharge rpm is fixed, which corresponds to a constant energy input and the development of specific temperature profile during the discharge.

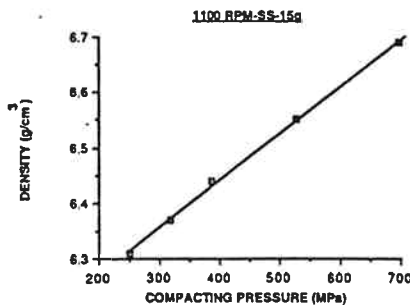


Figure 3 Compacting pressure effect on the compact final density.

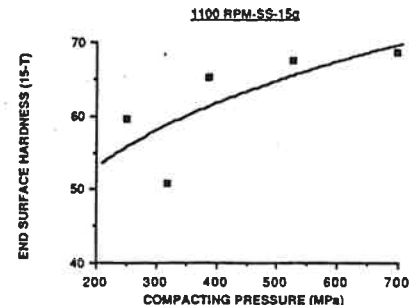


Figure 4 Compacting pressure effect on the compact end surface hardness.

*Effect of Electrode Materials:* Most experiments were done with stainless steel electrodes rather than copper electrodes. This is based on the fact that when all other process parameters are the same, stainless steel electrodes produce a denser compact than copper electrodes. Several factors account for this: a) stainless steel can sustain a much higher compacting pressure without any noticeable plastic deformation; b) while stainless steel consumes a large portion of the input energy due to its large electrical resistivity, the temperature rise of the electrodes caused by Joule heating has a very significant influence on the heat transfer at the electrode-compact interface, establishing a transient heat barrier for the compact during densification; c) the total amount of energy delivered to the load is higher in the case of stainless steel because the lower energy loss at brush-rotor interface associated with the smaller peak current and longer pulse length makes the energy conversion more efficient; d) the adhesion between the densified copper-graphite compact and the electrode is reduced; e) the electrodes are generally reusable.

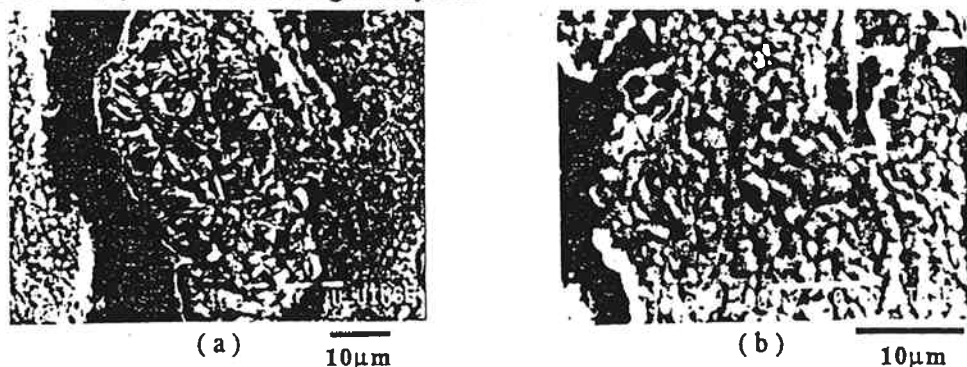


Figure 5 (a) Microstructure of the compact with intermediate energy input. (density = 99.85%, compacting pressure = 689 MPa) (b) close-up of the copper matrix

*Microstructures and Resistivity:* All the consolidated compacts were sectioned in the radial direction, metallographically prepared, and then examined by SEM. In general, a fairly uniform microstructure throughout the entire cross-section was observed. Localized recrystallization of copper matrix at the copper-graphite interface was seen in the relatively high density compacts. Dynamic recrystallization seems to have occurred during the process. Densification occurs in the copper matrix by effectively closing the internal and inter-particle pores. Excellent conformity between copper and graphite indicates an improved interfacial strength, as shown in figure 5. The microstructure in the densified composite material indicates the success of this rapid consolidation process in retaining very fine microstructure.



Electrical resistivity in the axial direction is plotted against compact density in figure 6. The theoretical value of electrical resistivity for copper-11 w/o graphite is calculated based on the law of mixtures. An order of magnitude decrease in resistivity was attained through the HEHR consolidation. When the electrical conductivity is plotted against the compact density, it is found that up to  $6.43 \text{ g/cm}^3$ , conductivity increases linearly with density. A similar relationship was observed by Taubenblatt in sintered high conductivity copper compacts[16]. The increase of resistivity in the region above  $6.43 \text{ g/cm}^3$  most likely results from the defects (high dislocation density) associated with large plastic deformation in the continuously cooled compacts. A comparison of the measured resistivity to the theoretical value indicates that a continuous copper matrix was maintained and a porosity of about 10% was effectively eliminated within 3 seconds by the HEHR consolidation. A projected low resistivity for copper-11 w/o graphite composite made from sintered copper-coated graphite [17] has been achieved by using HEHR consolidation. The reference commercial composite which is produced from similar powder stock has a resistivity of  $37 \mu\Omega\text{-cm}$ , with a density of  $5.5 \text{ g/cm}^3$ , as indicated by the arrow in figure 6.

**Nature of Pulsed HPG Heating:** In order to explore the effectiveness of the pulsed Joule heating, which is closely related to the mechanism of HEHR consolidation, computer simulation of heat transfer during the process was attempted. An explicit finite difference method was taken to solve the Fourier equation for an axisymmetric system. Actual dimensions and power input were used. Calculation stability was maintained with a time increment of 1 millisecond. The nature of rapid continuous heating and cooling can be easily seen in figure 7. The heating rate is around  $5 \times 10^4 \text{ K/s}$ , and the cooling rate is in the vicinity of  $500 \text{ K/s}$ . It should be noted that the total resistance which decreases rapidly from the beginning reaches the minimum as the temperature of the compact rises to the peak,

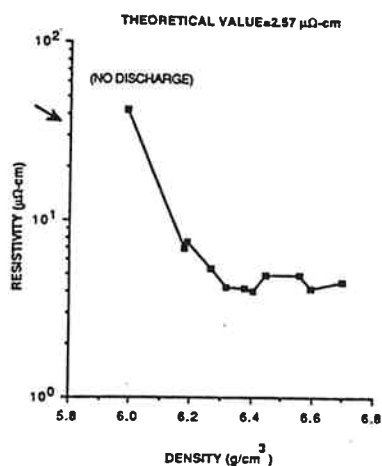


Figure 6 Relation between compact density and axial electrical resistivity.

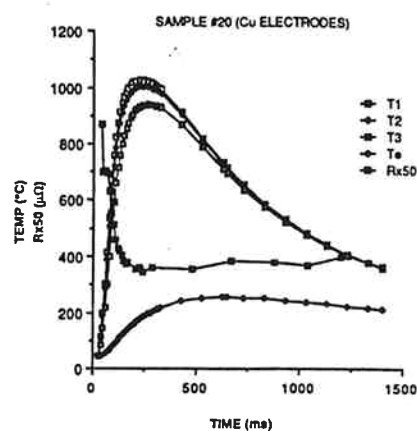


Figure 7 Simulated temperature change with time during powder consolidation process.

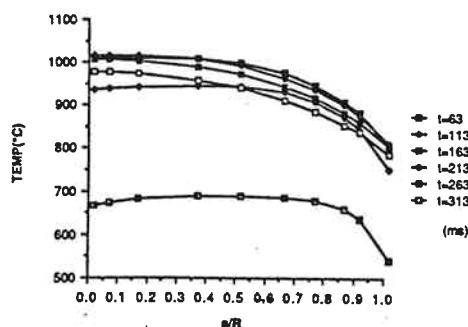


Figure 8 Simulated change of temperature distribution with time on the mid-plane of the compact.

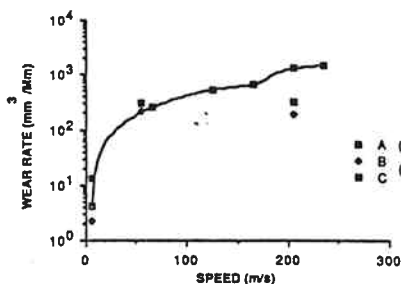


Figure 9 Comparison of wear behavior up to the sliding speed of 200 m/s.

indicating the main densification can proceed only as the temperature continues to increase. The same phenomenon was observed in the sintering of fine thoria powder under continuous heating, and it was concluded that plastic flow was the main densification mechanism[18]. The resistance rise after it reaches the minimum can be attributed to the effect of temperature rise in the electrodes. Figure 8 shows the change of temperature distribution on the mid-plane of the compact with time. The highest temperature quickly moves towards the center as the temperature increases. The center continues then to exhibit the highest temperature until the end of the discharge.

**Tribological Evaluation:** Preliminary evaluation of brush performance-related properties was accomplished by using testers with a wide range of sliding speeds (0-200 m/s). The HEHR consolidated composites of two different densities, i.e.,  $6.43 \text{ g/cm}^3$  (B) and  $6.58 \text{ g/cm}^3$  (C), along with the commercial P/M composite (A) were tested.

(1) Pulsed HPG system tester : The pulsed HPG system tester was designed as a test bed for HPG components. It allows simulation of the operating environment of HPG brushes. Voltage drop, current and temperature were measured. The composite brushes were discharged at 1000, 1200, and 2400 rpm in air without external cooling. For a 2400 rpm discharge, the brush sliding speed is 80 m/s. Table 2 shows the test results obtained for 3 discharges. For the HEHR consolidated brushes, no appreciable wear was found when all the tests were finished. A high current density (up to 14 MA/m<sup>2</sup>) capability at high sliding speeds was confirmed. A consistent low voltage drop and low contact resistance were observed at brush B.

Table 2

HIGH SPEED BRUSH TEST ON HPG SYSTEM TESTER									
BRUSH DOWN FORCE = 31.2 N									
BRUSH FACE AREA= 2.4 cm <sup>2</sup>									
DISCHARGE RPM	1000			1200			1200		
PULSE LENGTH (S)	5			3			3		
MACHINE VOLTS (V)	2.7			2.2			2.35		
MACHINE CURR (kA)	25			29			31		
TYPE OF BRUSH	A	B	C	A	B	C	A	B	C
BRUSH VOLTAGE	2.4	2.9	1.3	.55	.4	.55	.37	.18	.28
DROP (V)									
BRUSH CURR (kA)	--	--	--	2.8	3.4	2.04	--	--	--
TEMP (°C)	--	--	--	53	82	--	97	--	84

(2) High-speed brush tester : The high speed brush tester was mainly designed for investigation on flash temperature related phenomena. A thermal video imaging system was incorporated with the tester for real time temperature imaging. Continuous testing at speeds of up to 200 m/s at 6000 A is possible. The HEHR consolidated composite brushes were tested at 50 m/s for 80 minutes and at 200 m/s for 1 minute at zero current. In figure 9, the wear rates obtained from these tests were plotted with those for the reference composite studied by Casstevens et al.[19]. The wear rates for these three composites obtained from a pin-on-disk tester at 0.6 m/s are also plotted in this figure. Even though the load/area ratio is higher in the case of brushes B and C, their wear properties are quite comparable to the reference material A. As the speed goes up to 200 m/s, brushes B and C show only a slight increase over their wear rate at 50 m/s. However, the wear rate of the reference material A shows a linear increase with sliding speed, and exhibits approximately one order of magnitude increase in wear rate as speed increases from 50 m/s to 200 m/s. The HEHR consolidated copper-graphite composites clearly demonstrate their superior wear properties at high speeds.

#### ACKNOWLEDGMENT

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