

Comparison of thermal and microwave-assisted plasma sintering of nickel-diamond composites

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Abstract

There is considerable interest in processing technologies which can lead to more energy efficient sintering of metal powders. Microwave sintering has recently been shown to reduce energy usage as the volumetric heating process is considerably more efficient than resistance heating. RF plasma sintering meanwhile has been shown to deliver heat via uniform excitation of the processing gas resulting in ion bombardment of the workpiece. In this study the use of a rapid, novel microwave-assisted plasma sintering (MaPS) technology for processing of nickel-diamond metal matrix composites is evaluated. Nickel powder and polycrystalline diamond were mixed to prepare 20 mm discs under uniaxial compaction pressures of 100, 200 and 300 MPa. The discs were fired in a low pressure microwave plasma under a hydrogen atmosphere. For comparison, discs were also sintered using conventional tube furnace firing. The MaPS sintering is very rapid with full disc strength of >1000N, based on 3-point bend tests, being achieved within 10 minutes compared with 8 hours for furnace treatment. This study demonstrates that the microwave-assisted plasma sintered discs produced similar or superior performance to discs fired using furnace firing conditions but with sintering cycle time reduced by up to 95%.

Introduction

Plasma sintering has been demonstrated as a rapid and uniform sintering technique since its first reported studies by Bennett et al. [1]. Sintering has been carried out in plasmas generated using microwave, rf or DC power supplies [1-3] and the dominant heating mechanism is associated with ion and fast neutral bombardment. Diatomic gases such as hydrogen or nitrogen are typically used for plasma sintering. Plasma sintering is carried out under vacuum although unlike spark plasma sintering (SPS), there is no external force exerted on the workpiece [4]. There have also been a large number of reports in the literature on the microwave interaction with materials including the type of processing microwave heating can offer [5, 6]. The ability to penetrate the surface of the workpiece enables rapid volumetric heating in microwave processing, reducing the need for external heat sources [6]. Advantages of non-plasma microwave processing over furnace treatment include finer grain sizes, rounded porosity and higher ductility and toughness [7]. Increased shrinkage rates and decreased grain sizes can be achieved with microwave and plasma processing compared with furnace sintering [1, 8, 9]. With these advantages and significantly reduced cycle times, microwave-assisted plasma sintering (MaPS) offers an alternative route to production and reduced energy consumption per process.

This study compares, for the first time, furnace and microwave assisted plasma sintering of diamond metal matrix composites (MMCs). Nickel powder was chosen as the test material as it is used in a wide range of engineering components due to its corrosion resistance, wear resistance, mechanical strength, thermal expansion,

electrical conductivity and magnetic permeability [10]. Nickel powder is also used as a metal binder for many bonding applications in diamond and carbide machining tools [11].

Experimental Methods

Nickel-diamond MMCs were pressed in a uniaxial 20 mm diameter die at pressures of 100, 200 and 300 MPa (average green densities were 52, 58 and 62 % respectively). INCO nickel (T110) and nickel coated diamond (6-12 μm) from Element Six were mixed by weights of 80 and 20 % respectively in a T2F-Turbula for 100 minutes prior to pressing. Density was determined using the Archimedes principle in mercury and confirmed by measuring volume based on sample dimensions.

The microwave assisted plasma sintering process was carried out using a Circumferential Antenna Plasma (CAP) microwave system described in more detail elsewhere [12]. The plasma was formed at a pressure of 20 mbar in a hydrogen atmosphere. Three disc samples were rotated in a plasma located at the centre of the chamber per run (see Figure 1). Input powers of 2.4 kW were supplied from a Mugge microwave power supply operating at 2.45 GHz. Sample temperatures were measured using a LASCON QP003 two-colour pyrometer from Dr. Mergenthaler GmbH & Co. The use of a two-colour pyrometer is expected to eliminate the interference effect of the plasma on the emissivity of the sample. Typical heating and cooling rates of 7°C/s were observed in the microwave plasma fired samples. Emission spectroscopy was also carried out on the plasma using an Ocean Optics USB4000 spectrometer. Measurements were carried out to investigate changes in the electron density and total light emission of the plasma with increasing applied power. Furnace sintered samples were treated in tube furnace at 850°C in a flowing argon atmosphere. Heating rates of 4°C/min were used with the same dwell time of 10 minutes at the maximum temperature. The cooling rate was typically 2-3°C/min.

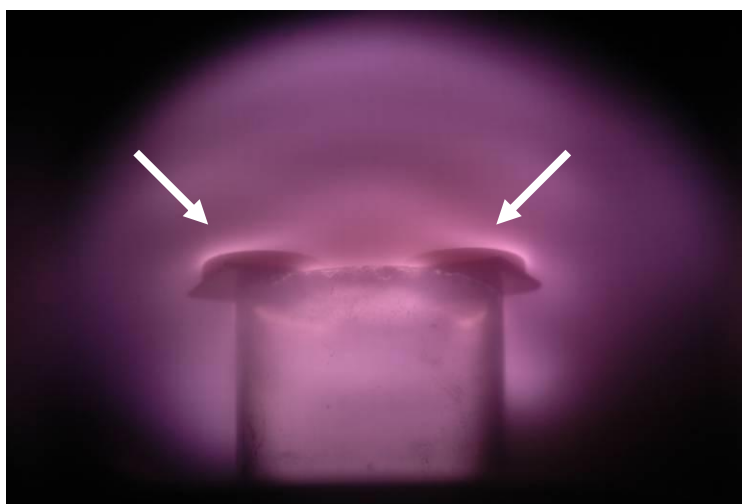


Figure 1: Samples (two visible) being sintered in a hydrogen microwave plasma

Flexural stress tests were carried out using a three point bend test. The span between the bottom pins (11 mm) and the cross sectional area of the fractured samples were used to calculate approximate flexural stress with an average value of three samples taken. Hardness testing was carried out using a Rockwell diamond indenter (HR 15 N scale), with the average value of 12 measurements taken. Rockwell indentation was

chosen as the large indent area (≈ 1 mm in diameter) should limit the effect of porosity or diamond reinforcement (≤ 20 μm). Pin-on-disc testing was undertaken using a TEER coatings POD-2 tester, with a 5 mm tungsten carbide ball as the pin, to evaluate the wear and tribological behaviour of the sintered samples [13]. The pin was loaded against the treated samples at 10 N at a linear speed of 5 cm/s for 1200 revolutions.

Results and discussion

Sintering conditions

Nickel-diamond samples pressed at 100, 200 and 300 MPa were sintered at 2.4 kW applied power in the microwave assisted hydrogen plasma. Following a two minute ramp up to an applied power of 2.4 kW, a maximum firing temperature of 850°C was achieved. Samples were sintered in sets of three with a 10 minute dwell time. A total cycle time of 20 minutes including pump down, firing and cooling was required. An increase in applied power resulted in an increase in plasma emission intensity (Figure 2). The peak shift and full width half maximum (FWHM) of the H_α and H_β , at 656.3 and 486.1 nm respectively, indicate the electron density in the plasma [14]. As the applied power increases from 1.2 to 2.4 kW, no increase in the normalised FWHM was observed and the ratio of the two peaks (H_α / H_β) remained almost constant at ≈ 5.3 . This indicates there is little or no increase in electron density with increasing applied power although the overall emission doubled as a result of a volume increase of the plasma. Furnace sintered samples were treated in a tube furnace at 850°C in batches of three in a flowing argon atmosphere. A total cycle time of over 8 hours, including heat up and cool down times, was observed.

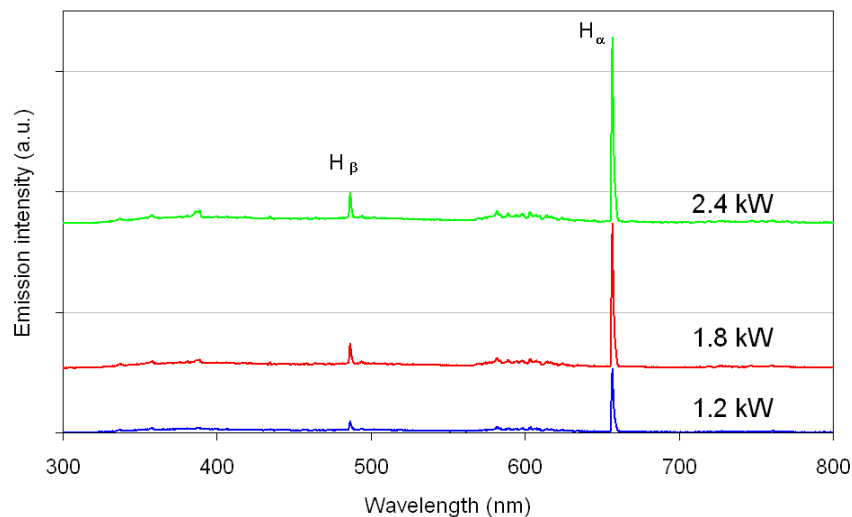


Figure 2: Emission intensity observed in hydrogen plasma

Mechanical behaviour of sintered MMCs

Density measurements were carried out using the Archimedes principle and also calculated based on the sample dimensions. As illustrated in Figure 3, there is little difference in sample density from the two sintering techniques despite a significant difference in total treatment times (20 minutes versus 8 hours). Sintering at these

temperatures resulted in a 15 % density increase for both plasma and furnace treated samples.

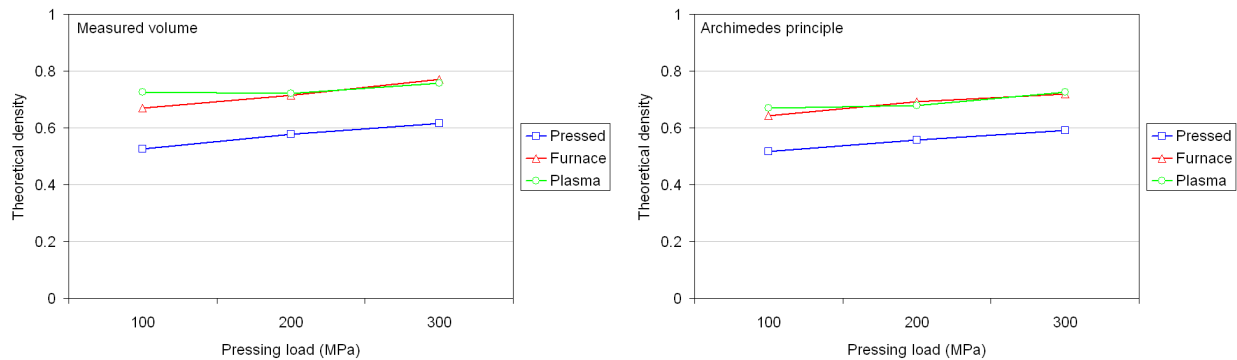


Figure 3: Density of pressed and sintered samples

Flexural stress values (see Figure 4) were determined using a three-point bend test. Due to the similarity in the cross-sectional area and densities achieved between the both sintering techniques, it was not unexpected that there was little difference observed between the two techniques. Samples pressed at higher pressures (300 MPa) performed better when microwave plasma treated and samples pressed at lower pressures (100 and 200 MPa) performing better when furnace treated.

As shown in Figure 4 however, microwave plasma fired samples exhibited significantly better hardness ($\geq 34\%$) than the furnace fired samples. As previously reported for microwave treated samples versus furnace treated samples, thermal equilibrium occurs almost instantaneously enabling sintering to occur in minutes [15].

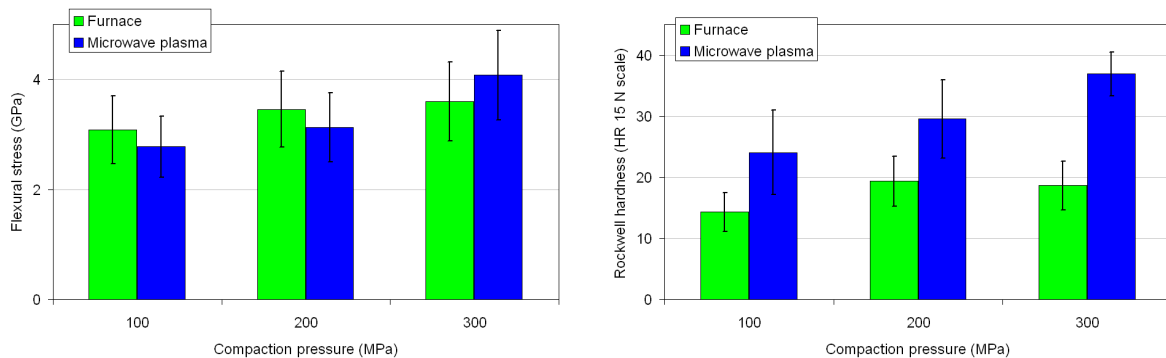


Figure 4: Flexural stress (left) and hardness values (right) of microwave assisted plasma and furnace sintered samples pressed at 100, 200 and 300 MPa

Examining the cross-sectional area of the nickel-diamond composites (Figure 5), it can be seen that the MaPS treated samples exhibit a much finer microstructure with increased nickel bonding between diamond particles (black areas) rather than increased flow into larger masses. The formation of larger masses of nickel may lead to larger pores in the furnace treated samples resulting in reduced hardness. This is most likely due to the heating rate of $\approx 420^\circ\text{C}/\text{min}$ versus $4^\circ\text{C}/\text{min}$ for MaPS over furnace treatment. These heating and equivalent cooling rates can be achieved as the high energy plasma discharge is isolated from the colder surroundings ($\approx 30^\circ\text{C}$) of the discharge chamber. The finer microstructure of the MaPS treated samples may explain the increased hardness values observed [7].

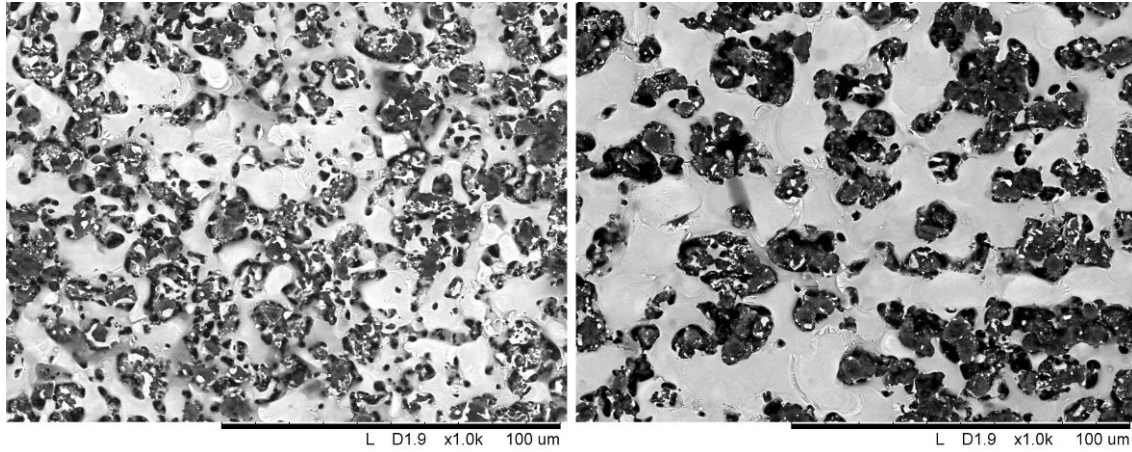


Figure 5: Polished cross-sectional microstructure of 300MPa pressed MaPS (left) and furnace (right) sintered

Pin on disc wear testing was undertaken on each of the samples to determine the wear performance of samples pressed at increasing pressures. The wear track generated was examined using SEM. The wear track itself appeared to be masked in a foreign material, as shown in Figure 6. Using EDX elemental analysis a large presence of tungsten in the wear track was detected with a maximum concentration of 67 % in wear track centre. This result demonstrated that there was significant wear of the tungsten carbide pin. Due to this wear of the pin, direct comparison of the wear tracks was not possible.

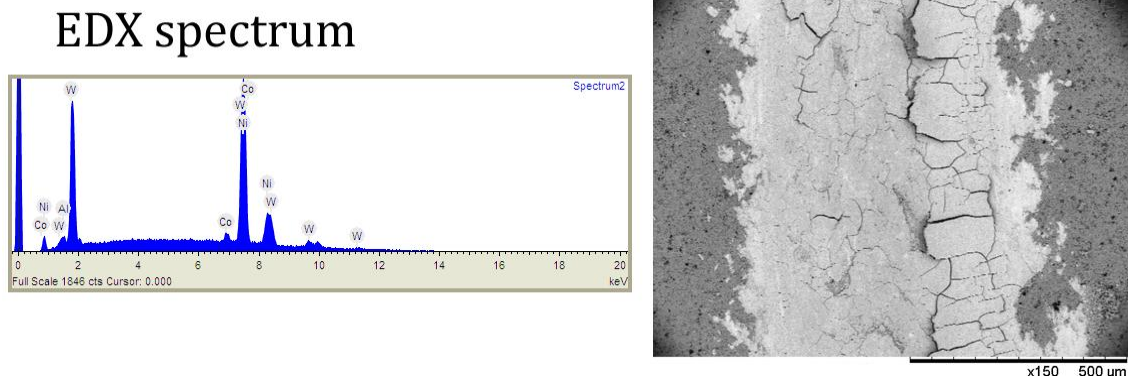


Figure 6: EDX analysis (left) and SEM image (right) of pin-on-disc wear track

The wear rate of the pin was measured using an optical microscope and ImageJ analysis software [16]. For each of the sample compaction pressures (100, 200 and 300 MPa), it was observed that microwave plasma sintered samples caused an increased level of pin wear compared to furnace sintered samples (3, 19 and 41 % increase respectively). This is not unexpected given the increased surface hardness of plasma sintered samples. However, samples pressed at highest pressures caused lower levels of pin wear compared with those pressed at the lower pressures for both firing techniques. This may be due to increased diamond pull out due to the decreased density of the 100 MPa pressed samples. As the diamond is removed, an increased level of abrasion may occur due to the abrasive debris rather than the diamond containing MMC disk. To examine this requires further testing using a different pin materials or disc-on-disc wear testing.

Conclusions

Nickel-diamond MMCs have been produced using microwave assisted plasma sintering (MaPS) and compared with composites produced using furnace sintering. As the plasma is isolated from the 'colder' walls of the vacuum chamber, rapid heat up and quenching rates can be achieved resulting in a treatment time reduction of 95%. Despite the significantly reduced treatment times comparable sample densities were observed in samples from the two techniques. Similar breaking strengths were also observed for both furnace and plasma fired MMCs although plasma sintered samples exhibited increased hardness values, most likely due to the finer microstructure of the MaPS samples. MaPS treated MMCs caused greater tungsten carbide pin wear during pin-on-disc testing, while samples pressed at lower pressures tended to exhibit higher pin wear rates.

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