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Effect of Ni/C on the mechanical properties and microstructure of synthetic diamond brazed with Ni-Cr brazing filler metal

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ABSTRACT

Ni-Cr brazing filler metal containing varying amounts (0 wt%, 3 wt%, and 5 wt%) of nickel-coated graphite (Ni/C) was employed to prepare brazed synthetic diamond samples. The interfacial microstructure characterization and thermal damage of brazed synthetic diamond were investigated using scanning electron microscope and Raman spectroscopy. The mechanical properties of static pressure strength and impact toughness of brazed synthetic diamond were investigated in accordance with the machinery industry standard. The hardness of brazed layer was measured using a micro-hardness tester. The results demonstrate the presence of lamellar compounds Cr_3C_2 and void columnar compounds Cr_7C_3, as well as filamentous eutectic graphite, on the surface of brazed synthetic diamond following aqua regia corrosion. Increasing the Ni/C ratio in the Ni-Cr brazing filler metal leads to a reduction in graphitization degree of synthetic diamond. When the Ni-Cr brazing filler metal contains 5 wt% Ni/C, the compressive stress in brazed synthetic diamond is reduced by 65 MPa, the static pressure strength and impact toughness of brazed synthetic diamond are increased by 15.1% and 29.9%, respectively, and the hardness of brazed layer is decreased by 79 HV, this is beneficial to enhance self-sharpness of synthetic diamond tools brazed with Ni-Cr brazing filler metal.

Keywords: Ni/C; Microstructure; Mechanical properties; Synthetic diamond; Ni-Cr brazing filler metal.

1. INTRODUCTION

Due to the exceptional mechanical and chemical properties, such as impressive strength, unparalleled hardness, and remarkable chemical stability, synthetic diamond has found extensive applications in the field of cutting and grinding processing since it was successfully synthesized in 1955 [1–3]. Brazing is considered one of the most promising bonding technologies for synthetic diamond, as it enables chemical and metallurgical bonding between synthetic diamond, brazing filler metal, and the tool substrate at the selected temperatures [4]. Synthetic diamond's high chemical inertia necessitates the use of brazing filler metal containing reactive elements such as Ti, Cr, V, or W to form chemical compounds at the interface with synthetic diamond during brazing, distinguishing it from electroplating and sintering [5–7]. Nowadays, Ag-Cu-Ti alloy [8, 9], Cu-Sn-Ti alloy [10, 11], and Ni-Cr alloy [12, 13] are widely utilized as brazing filler metal. Ni-Cr alloy possesses high strength, wear resistance, and good wettability to synthetic diamond. Additionally, the presence of element Cr allows for a reaction with synthetic diamond, forming reactants Cr_3C_2 and Cr_7C_3 which ensure a high bonding strength between synthetic diamond and Ni-Cr alloy [13]. As a result, Ni-Cr alloy has become one of the most prevalent choices for preparing brazed synthetic diamond tools. However, despite its numerous advantages mentioned above, Ni-Cr alloy also presents certain deficiencies such as requiring high brazing temperature and containing graphitizing elements Ni and Fe that can lead to the graphitization of synthetic diamond during brazing [14, 15]. After brazing, significant residual stress remains in the interior of synthetic diamond due to the difference in thermal expansion coefficients between Ni-Cr alloy and synthetic diamond. This can lead to degradation of the mechanical properties of synthetic diamond [16–18]. The high wear resistance of Ni-Cr alloy also prevents it from maintaining a proper grinding ratio with synthetic diamond during grinding, ultimately reducing the service life of brazed synthetic diamond tools [19–21]. Therefore, it is very important to study the method which can reduce the thermal damage of synthetic diamond brazed with Ni-Cr brazing filler metal and improve the self-sharpness of brazes synthetic diamond tools.

Scholars conducted constructive studies to address the issue of thermal damage and proper grinding ratio for brazed synthetic diamond tools. The research indicates that brazing diamond with Ni-Cr composite alloy

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containing 1 wt % multi-layer graphite resulted in a reduction of thermal damage, a 6.6% decrease in residual stress of brazed diamonds, and a 9% increase in static pressure strength and 7.9% increase in toughness against impact [22, 23]. The compressive residual stress value of brazed diamond grits is –1.3 GPa, large residual stress will cause cracks in the synthetic diamond. It exhibited the longest tool life, highest drilling efficiency, and mildest wear modes, when a brazed diamond drill bit was fabricated from Ni-Cr composite alloys including 1 wt% multilayer graphite addition. Nevertheless, during the drilling operation, excessive multi-layer graphite will cause early shedding of synthetic diamond from the Ni-Cr brazing filler metal during the drilling process [24]. When Ni-Cr brazing filler metal added with 5 wt% Cu-Ce alloy were used to braze synthetic diamond segments, the highest resistance, machining performance, and the best surface morphology were obtained, however, excessive Cu-Ce alloy will cause a rapid decline in wear resistance [25–27].

Graphite has exceptional properties, such as a low coefficient of thermal expansion, high absorption of Ni metal atoms, and excellent thermal conductivity, so it may be able to mitigate the thermal damage (graphitization, residual stress, and chemical attack) to synthetic diamonds brazed with Ni-Cr brazing filler metal. Because graphite has a relatively low density, it is not easily blended with Ni-Cr brazing filler metal. Furthermore, the studies indicate that graphite is likely to accumulate at the top layer of the brazing filler metal layer following brazing.

In order to solve the key problem of synthetic diamond brazed with Ni-Cr brazing filler metal and the insufficiency of adding graphite as composite brazing filler metal, in this work, brazed diamond samples were created using various Ni/C (0 wt%, 3 wt%, and 5 wt%) reinforced Ni-Cr brazing filler metal compositions that were developed. The interfacial microstructure characterization between synthetic diamond and Ni-Cr brazing filler was analyzed. The mechanical properties of brazed synthetic diamond (graphitization degree, residual stress distribution, static pressure strength, and the impact toughness) and the hardness distribution of brazing layer were measured.

2. MATERIALS AND METHODS

2.1. Materials and brazing procedure

The synthetic diamond was purchased from Zhongnan Diamond Co. Ltd., and the particle size is 30/35 mesh (500~600 µm). The size of Ni/C particles ranges from 80 to 100 µm. Figure 1a and 1b show the morphology of synthetic diamond and Ni/C, respectively. The synthetic diamond has a mesh size of 30/35 and a size of 500~600 µm, Ni/C particles range in size from 80~150 µm. Ni-Cr alloy (from the Beijing General Research Institute of Mining and Metallurgy) was used as the brazing filler metal. The specific composition of Ni-Cr brazing filler metal is shown in Table 1. The tool substrate was Q235A (chemical elements: C 0.2%, Mn 0.45%, S 0.05%, P 0.04%, Si 0.3%) with a diameter of 20 mm and a height of 10 mm.

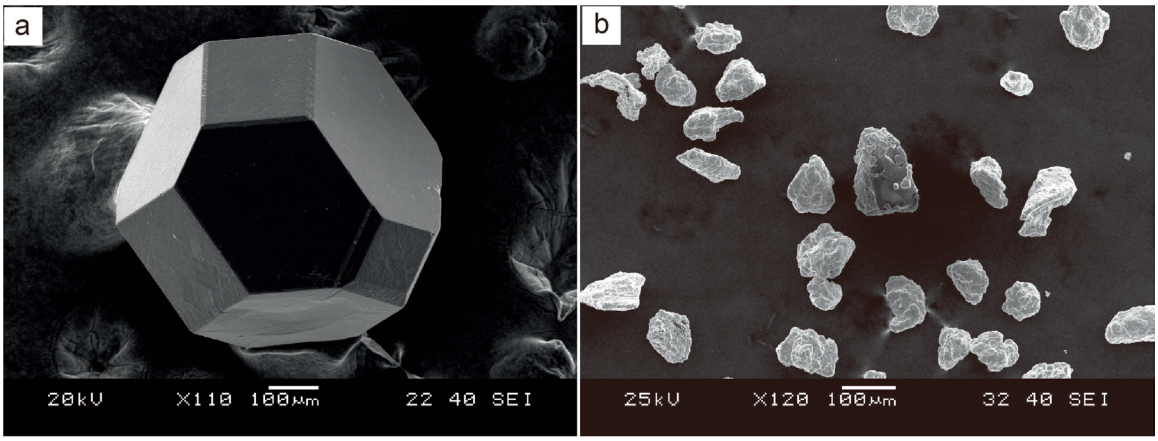

Figure 1: Morphology of synthetic diamond (a) and Ni/C (b).

Table 1: Chemical composition of Ni-Cr brazing filler metal.

ELEMENT	Cr	C	B	Si	Fe	Ni
Mass fraction	16~18	0.6~0.8	2.5~3.0	4.0~5.0	5	balance

Before brazing, strict cleaning was taken on all the materials to remove the impurities and obtain a good brazing interface among synthetic diamond, Ni-Cr brazing filler metal, and tool substrate Q235A. First, the surface of the Q235A was polished with grinding wheels to remove the rust. Next, synthetic diamond and Q235A were cleaned in acetone with ultrasonic agitation for 10 minutes, respectively, and dried in air. Then the Ni-Cr brazing filler metal added with Ni/C (mixing uniformity by mechanical method, 0 wt%, 3 wt%, and 5 wt%) was sprayed uniformly on the surface of Q235A; subsequently, synthetic diamond grains were placed onto the surface of Ni-Cr brazing filler metal. Finally, the specimen was inserted into the protective atmosphere furnace, and Argon was utilized as the protective atmosphere. Brazing was carried out at 1050 °C with a holding time of 5 minutes. The heating rate is kept at 10 °C/min. The sample is cooled in the furnace. The samples were taken out of the protective atmosphere furnace when all the brazing was complete.

2.2. Characterization methods

The morphology of brazed synthetic diamond samples and the interface microstructure between synthetic diamond and Ni-Cr brazing filler metal were examined by a scanning electron microscope (JSM-6300) accompanied by energy dispersive spectrometry (EDS). After the Ni-Cr brazing filler metal and carbide on the surface of synthetic diamond were removed by the electrolysis process with 10% dilute sulfuric acid, the graphitization and residual stress of the brazed synthetic diamond were studied by Raman spectroscopy (JY LabRAM HR Evolution) in the frequency range 1000 ~ 1700 cm^{-1}. The compressive strength and impact toughness of brazed synthetic diamond were studied by ZMC-II type compressive strength testers for single particles and CYCJ-04A type impact toughness testers for superhard abrasives according to Machinery Industry Standard of the People's Republic of China JB/T 7988.1-1999 and Machinery Industry Standard of the People's Republic of China JB/T 6571-93, respectively. The hardness of the brazed layer was measured by an HXD-1000 TC micro-hardness tester.

3. RESULTS AND DISCUSSION

3.1. Microstructure between synthetic diamond and Ni-Cr brazing filler metal

In order to study the interface microstructure between synthetic diamond and Ni-Cr brazing filler metal, the brazed synthetic diamond samples were corroded with aqua regia to remove the Q235 steel substrate and Ni-Cr brazing filler metal. Figure 2 displays the microstructure of brazed synthetic diamond after chemical etching. Several compounds form on the surface of brazed synthetic diamond. When the synthetic diamond is brazed with Ni-Cr brazing filler metal, the reaction products are mainly flake carbide A and cylindrical carbide B. When Ni/C is added to Ni-Cr brazing filler metal, it not only forms flake carbide A and cylindrical carbide B on the surface of brazed synthetic diamond, but also forms filamentous substances, meanwhile, the carbide B becomes hollow structure.

The chemical components of products A, B, and C are shown in Table 2. Combined with the reference [6] and [21], we can know that carbides A and B are Cr_3C_2 and Cr_7C_3, respectively. Compared with synthetic diamond brazed with Ni-Cr brazing filler metal, when synthetic diamond brazed with Ni-Cr brazing filler metal added Ni/C, the quantity of carbide Cr_3C_2 reduces, and the structure of Cr_7C_3 changes. This is related to the addition of Ni/C in Ni-Cr brazing filler metal. The amount of carbide Cr_3C_2 is reduced, which illustrates that the chemical reaction between synthetic diamond and Ni-Cr brazing filler metal is reduced. This will help improve the quality of brazed synthetic diamond. When synthetic diamond is brazed with Ni-Cr brazing filler metal added Ni/C, graphite can be used as the heterogeneous nucleated core for carbide Cr_7C_3. Carbide Cr_7C_3 nucleates on the surface of graphite with screw dislocation; this will cause the initially formed hexagonal lattice carbide Cr_7C_3 to grow in a spiral pattern. The low-melting-point liquid alloy is encircled with carbide Cr_7C_3, and spiral step could continue to grow by absorbing neighbor atoms. The core of the screw dislocation is separated from the boundaries. During the process of crystal growth by adsorbing the atom of the spiral step, the crystal center keeps hollow and is suffused with alloy liquid. Meanwhile, after brazing, the cooling of the brazed samples is faster for the protective atmosphere furnace, and the alloy has lower melting points than carbide Cr_7C_3. At last, it forms hollow Cr_7C_3.

In the middle of carbides Cr_7C_3, filamentous substances created when synthetic diamond brazed with Ni-Cr brazing filler metal added Ni/C. Carbon is the main component of filamentous substances, as indicated by the energy spectrum test finding for product C. Combined with the phase diagram of Nickel-Carbon, at high temperatures, the concentration of carbon in Ni-Cr brazing filler metal decreases with the decreasing brazing temperature; during the eutectic transformation, it forms eutectic graphite of fascicular filamentary, so we can assume that the filamentous substance C is eutectic graphite.

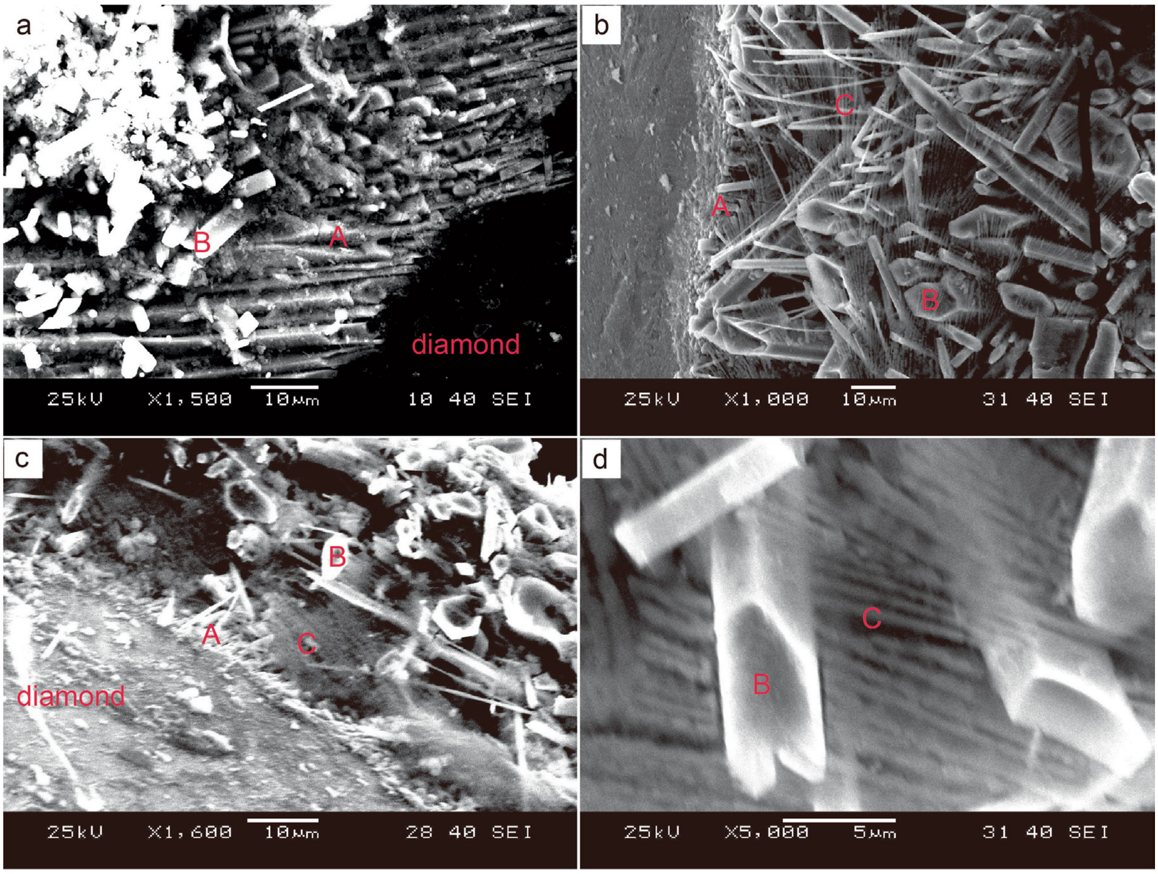

Figure 2: Microstructure of brazed synthetic diamond after chemical etching (a) 0 wt% Ni/C, (b) 3 wt% Ni/C, (c) 5 wt% Ni/C, (d) enlarged view of (b).

Table 2: Chemical composition of each point (atomic fraction) %.

LOCATION	ELEMENT			
	Cr	Ni	C	Si
A	58.7	0	41.5	0
B	67.8	0	32.2	0
C	0	0.5	98.2	0.7

3.2. Thermal damage analysis of synthetic diamond brazed with Ni-Cr brazing filler metal

Figure 3 shows the morphology of brazed synthetic diamond samples after electrolytic corrosion with 10% dilute sulfuric acid. Compared with Figure 1(a), the morphology of synthetic diamond has a complete morphology, and the edges are clearly visible. This illustrates that the degree of chemical erosion of synthetic diamond is relatively light when it is brazed with Ni-Cr brazing filler metal with the brazing process used in this work. As shown in Figure 3(b), the surface of brazed synthetic diamond is rough and uneven; some textures are distributed in a certain direction; this is the morphology retained by carbide Cr_3C_2 when it is corroded away. It also has a complete shape and clear edges when synthetic diamond is brazed with Ni-Cr brazing filler metal added Ni/C. The surface of synthetic diamond in Figures 3(c) and (d) is relatively flat compared with Figure 3(b), which means that the chemical erosion of synthetic diamond is lighter when it is brazed with Ni-Cr brazing filler metal added Ni/C.

The surface of diamond in Figure 1(a), Figure 3(a), (c) and (d) was analyzed with a Roman spectrometer, and the results of synthetic diamond brazed with different brazing filler metal are shown in Figure 4. In addition to the diamond peak of 1332 cm^{-1}, peaks 1358 cm^{-1} and 1585 cm^{-1} also appeared on the surface of diamond when synthetic diamond was brazed with Ni-Cr brazing filler metal. It is known that peaks 1358 cm^{-1} and 1585 cm^{-1} are graphite characteristic peaks and amorphous carbon peaks [14], respectively. This indicates that graphite and amorphous carbon formed on the surface of synthetic diamonds, after the synthetic diamond was

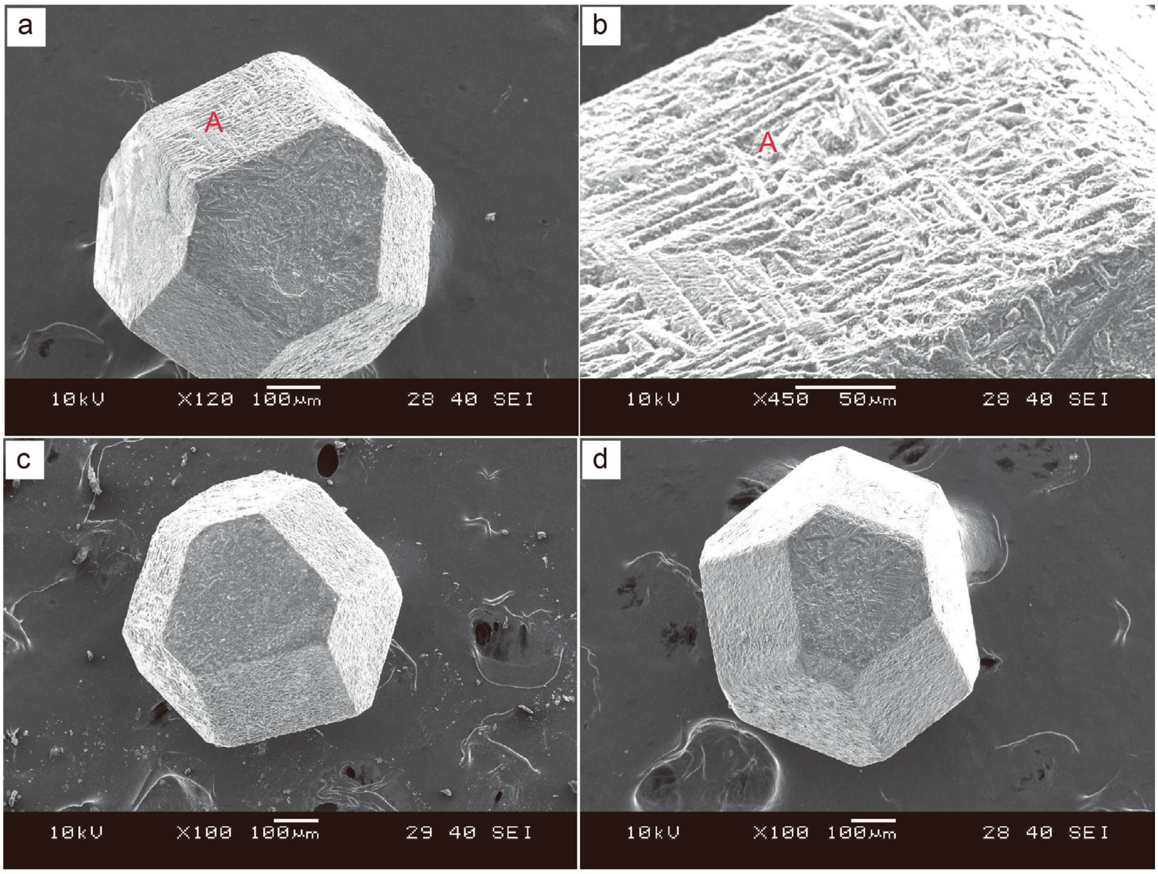

Figure 3: Morphology of brazed synthetic diamond after electrolytic corrosion (a) 0 wt% Ni/C, (b) enlarged view of (a), (c) 3 wt% Ni/C, (d) 5 wt% Ni/C.

brazed with Ni-Cr brazing filler metal. With the increase in Ni/C content in Ni-Cr brazing filler metal, the peak value of diamond increased, and the peaks of graphite and amorphous carbon decreased. This indicates that the degree of graphitization decreased when Ni-Cr brazing filler metal added Ni/C to braze synthetic diamond. The sensitivity of graphite to Raman is 50 times greater than that of diamond to Raman; this illustrates that the amount of graphite and amorphous carbon is very small; diamond is the main component. The synthetic diamond is only graphitized on the surface, not all of it. When synthetic diamond is brazed with Ni-Cr brazing filler metal without adding Ni/C, under the action of infiltration of Ni and Cr, diamond is translated into graphite by dissolving and diffusing C on the surface of synthetic diamond. According to the chemical reaction of synthetic diamond translating into graphite, it can be known that adding Ni/C can restrain the translation of synthetic diamond to graphite; at the same time, graphite can restrain the excessive interface reaction between synthetic diamond and Ni-Cr brazing filler metal, which is beneficial to reduce the performance degradation of synthetic diamond.

Due to the difference in elastic modulus, Poisson's ratio, and coefficient of thermal expansion among synthetic diamond, Ni-Cr brazing filler metal, and Q235 steel substrates, a large residual stress was formed after brazing. Residual stress is an important factor effecting the interface bonding strength between synthetic diamond and Ni-Cr brazing filler metal. When the residual stress is too large, it will not only lead to cracks in the synthetic diamond but also cause the premature loss of synthetic diamond from Ni-Cr brazing filler metal. The residual stress of brazed synthetic diamond was measured by the offset of the Raman stokes peak between brazed synthetic diamond and non-brazed synthetic diamond. Figure 5 shows the residual stress of synthetic diamonds brazed with different brazing filler metal. It has the same trend of residual stress in synthetic diamonds brazed with brazing filler metal. In the top region of synthetic diamond, it is under tensile stress. As the depth increases, the tensile stress is translated into compressive stress. After adding Ni/C to Ni-Cr brazing filler metal, the tensile stress of synthetic diamonds barely changed, but the compressive stress reduced; it changed from 618 MPa to 553 MPa. In the top region of brazed synthetic diamond, there is no Ni-Cr brazing filler metal; Ni-Cr brazing filler metal has a negligible effect on the synthetic diamond. The cooling rate is very small; it can form uniform temperature fields in brazed synthetic diamond, and the residual stress developing during the brazing process is

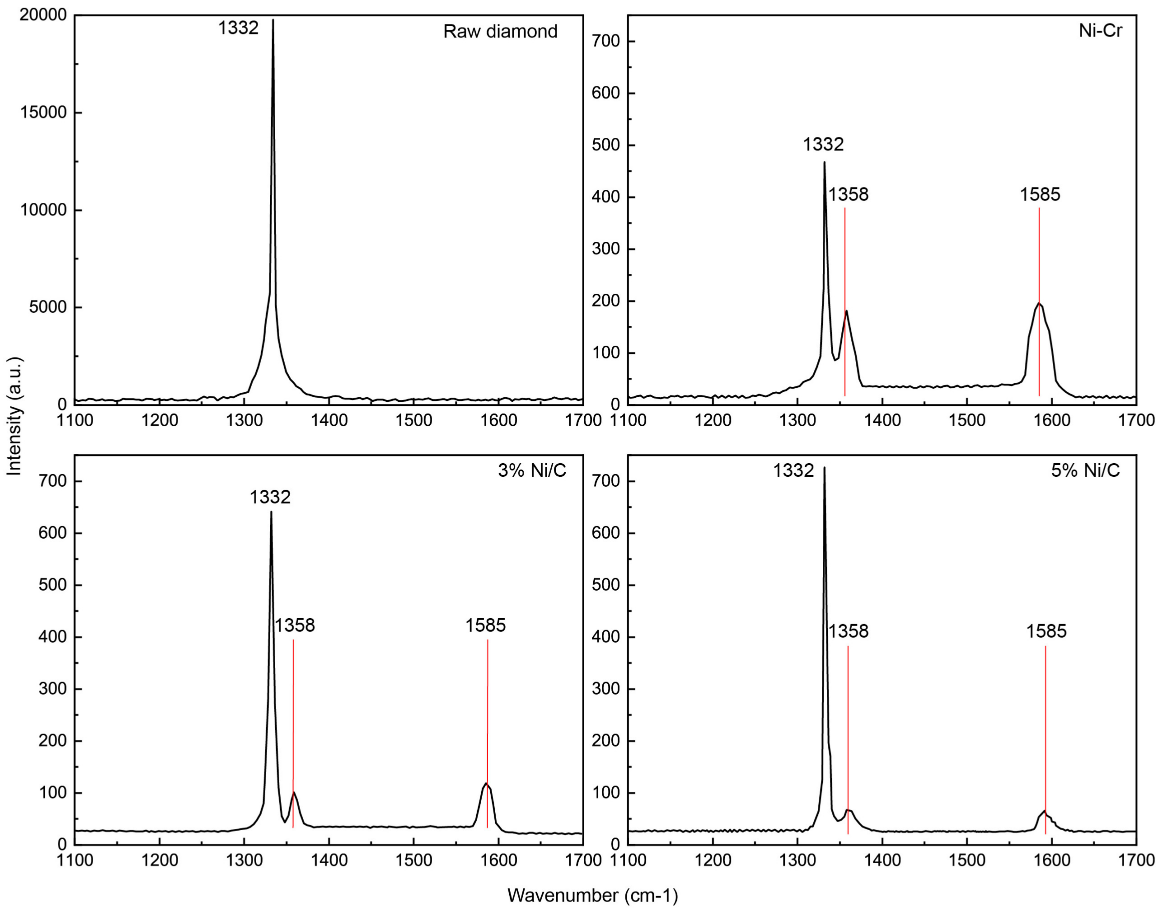

Figure 4: Raman spectroscopy results of synthetic diamond under different conditions.

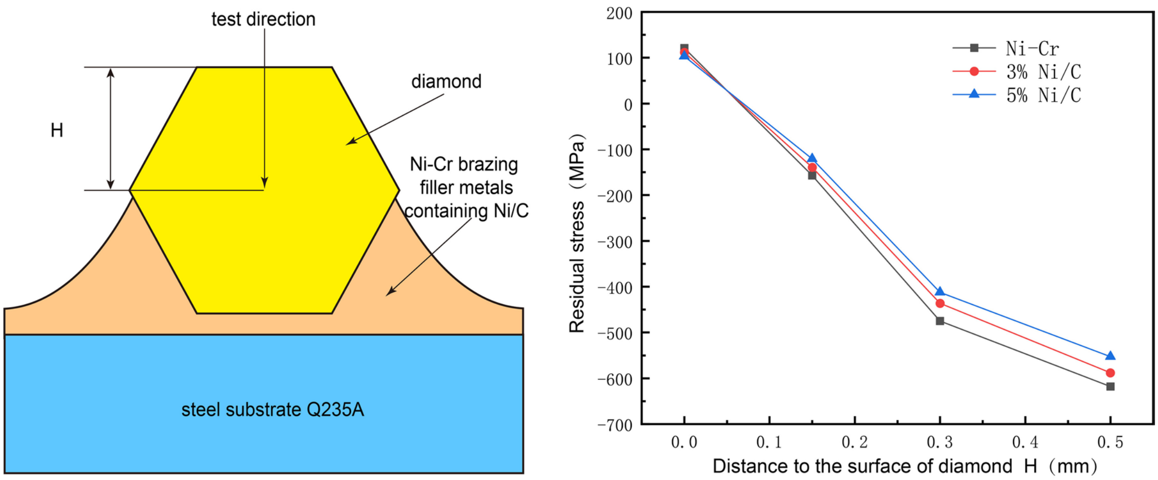

Figure 5: Residual stress of synthetic diamond brazed with different brazing filler metal.

small. With the depth of synthetic diamond in Ni-Cr brazing filler metal increasing and the large difference in thermal expansion coefficients and modulus of elastic between synthetic diamond and Ni-Cr brazing filler metal, the synthetic diamond was stressed by the shrinkage of the Ni-Cr brazing filler metal during cooling. The greater of depth of synthetic diamond in Ni-Cr brazing filler metal, the greater the compress stress forms in the brazed synthetic diamond. After adding Ni/C to Ni-Cr brazing filler metal, the Ni/C in the Ni-Cr brazing filler metal will act as soft particle to relieve stress, reducing the compress stress in the brazed synthetic diamond.

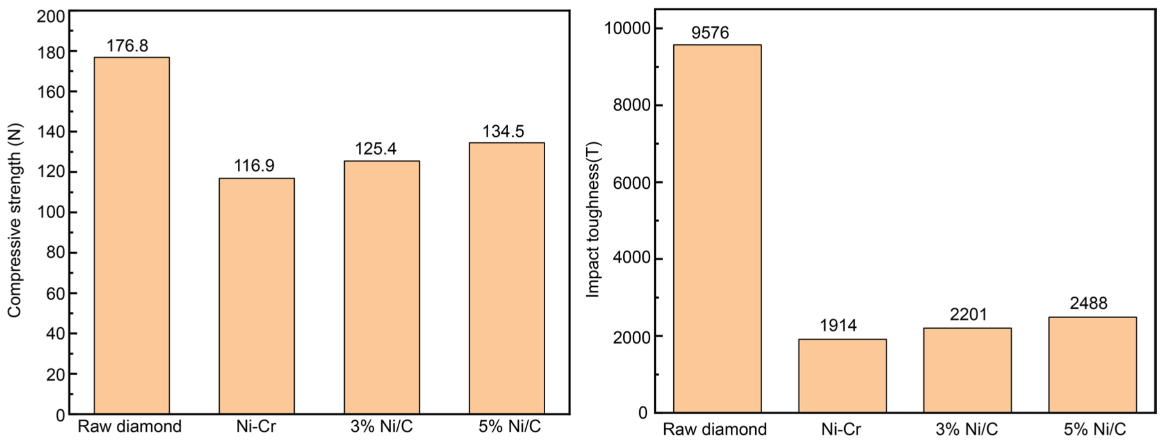

Figure 6: Compressive strength and impact toughness of synthetic diamond under different conditions.

3.3. Mechanical properties of synthetic diamond brazed with Ni-Cr brazing filler metal

The performance of brazed synthetic diamond tools depends not only on the bonding strength of synthetic diamond and brazing filler metal but also on the mechanical properties of synthetic diamond after brazing. Static pressure strength and impact toughness are very important parameters to characterize the mechanical properties of synthetic diamonds. Figure 6 shows the static pressure strength and impact toughness of synthetic diamond under different conditions. The static pressure strength of synthetic diamond decreases after brazing in a controlled atmosphere; this indicates that the brazing causes a certain degree of thermal damage to the synthetic diamond. This is largely due to the difference in coefficient of thermal expansion between the synthetic diamond and the trace impurity elements (Fe, Co, and Ni) in the synthetic diamond.

The thermal expansion coefficient of impurity elements is higher than that of synthetic diamond; this will cause the synthetic diamond to bear greater residual stress after brazing, and the synthetic diamond may crack. At the same time, the corrosion pits formed by chemical erosion on the surface of synthetic diamond reduce the flatness of the surface, which will result in a decrease in the static pressure strength of synthetic diamond. After adding Ni/C to Ni-Cr brazing filler metal, the residual stress formed in brazed synthetic diamond and the chemical erosion on the surface of synthetic diamond are reduced, which will improve the static pressure strength of synthetic diamond. The static pressure strength increased from 116.9 N to 134.5 N. The impact toughness of synthetic diamond under different conditions has the same change trend with static pressure strength; however, when synthetic diamond was brazed with a protective atmosphere, the impact toughness decreased very seriously, from 9576 to 1914. Chemical erosion and graphitization of synthetic diamonds will cause the surface coarsening of brazed synthetic diamonds, which results in the friction and adhesion of synthetic diamonds during impact, reducing the impact toughness of synthetic diamonds; meanwhile, residual stress will also further reduce the impact toughness of synthetic diamonds. After adding Ni/C to Ni-Cr brazing filler metal, the impact toughness of synthetic diamond has a certain degree of improvement, from 1914 T to 2488 T. This has a relation with the reduction of residual stress and chemical erosion.

3.4. Hardness distribution of brazed layer

The hardness of the brazed layer has a very important influence on the performance of brazed synthetic diamond tools. In the grinding process, only synthetic diamond has always maintained a high exposure height to make brazed synthetic diamond tools continue to maintain a high sharpness. If the hardness of the brazed layer is high, the grinding ratio of the synthetic diamond and brazed layer cannot be consistent, which will lead to a decrease in the exposure height of the synthetic diamond and affect the sharpness of the synthetic diamond tools. Due to the high hardness and wear resistance of Ni-Cr brazing filler metal, the sharpness of brazing synthetic diamond tools made of Ni-Cr brazing filler metal is not enough, and the grinding efficiency is reduced. Figure 7(a) shows the hardness distribution of the brazed layer, Figure 7(b) shows the location of the hardness tested in the direction along the line. The hardness of the brazed layer decreases gradually from the interface between synthetic diamond and Ni-Cr brazing filler metal to the Q235 steel substrate, regardless of whether Ni/C is added to the Ni-Cr brazing filler metal. When Ni/C is not added to the Ni-Cr brazing filler metal, the hardness of the brazed layer is about 820 HV with a small change range. After adding Ni/C to the Ni-Cr brazing filler metal, the hardness of the brazed layer is reduced to 741 HV-789 HV, although it has a certain range of change, but the change is small. The hardness of the brazed layer decreases with the increase in Ni/C content.

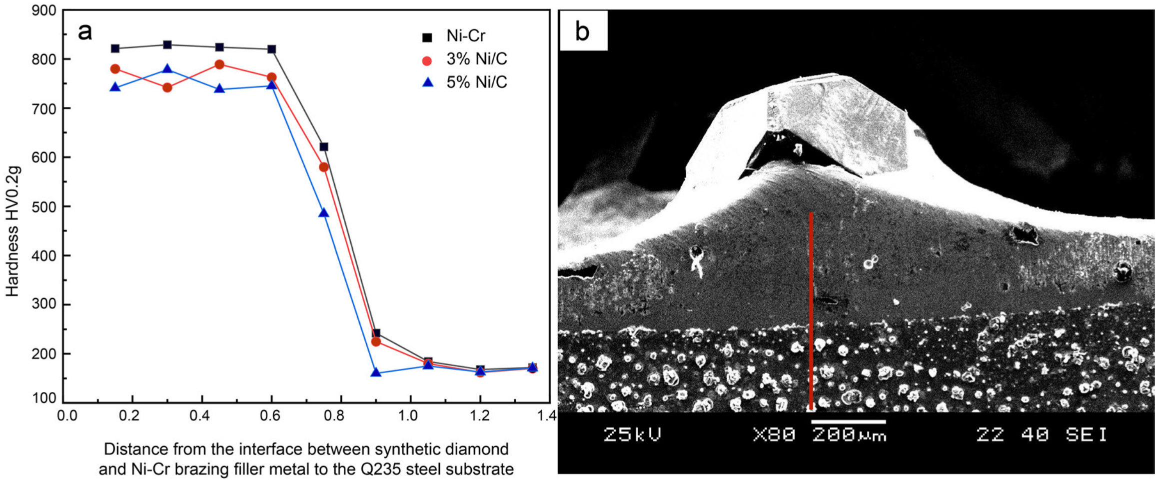

Figure 7: Hardness distribution of brazed layer (a) hardness distribution, (b) hardness test location.

When Ni-Cr brazing filler metal added with Ni/C was used to braze synthetic diamond, in the process of brazing, active elements such as Cr can form compounds with element carbon, which can improve the hardness of the brazed layer to a certain extent. After brazing, there is a certain distribution of carbon in the brazed layer; it exists as soft particles, which can reduce the hardness of the brazed layer. Under the combined action of the two, the hardness of brazed layer reduces to a certain extent, but the reduction is not large. The hardness of the brazed layer is reduced, which is beneficial to adjust the grinding ratio of synthetic diamond and brazed layer. The exposed height of synthetic diamond can be increased to a certain extent, which can ensure the synthetic diamond tools have better self-sharpness.

4. CONCLUSIONS

Compared with Ni-Cr brazing filler metal brazing synthetic diamond, when synthetic diamond is brazed with Ni-Cr brazing filler metal added with Ni/C(3 wt%, 5 wt%) under the same brazing process conditions, the following conclusions can be drawn.

On the surface of synthetic diamond, which is composed of lamellar compounds Cr_3C_2, void columnar compounds Cr_7C_3, and filamentous eutectic graphite. Both the amount of carbide Cr_3C_2 and the graphitization degree of synthetic diamond are decreased.

When the Ni/C content in Ni-Cr brazing filler metal is 5 wt%, in synthetic diamond, the compressive stress dropped from 618 MPa to 553 MPa. The static pressure strength and impact toughness of synthetic diamond increased from 116.9 N to 134.5 N, and 1914 T to 2488 T, respectively. The hardness of the brazed layer dropped from 820 HV to 741 HV, which is helpful to improve the self-sharpness of brazed synthetic diamond tools with Ni-Cr brazing filler metal.

5. ACKNOWLEDGEMENTS

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