TOPICAL REVIEW

Research status on surface metallization of diamond

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Abstract

With the highest hardness and excellent wear resistance, diamond is the most suitable abrasive for the manufacture of hard materials processing tools. It has been widely used in the manufacture of tools for cutting, grinding, polishing and dressing. However, there is high interfacial energy between diamond and binder, which reduces the service life and processing efficiency of diamond tools. Metallization of diamond surface is an effective way to improve the performance and quality of diamond tools, and has become a hot spot in the research field of diamond tools. Based on the related literatures at home and abroad, this paper reviews the plating technology of single metal coating, alloy coating and composite coating on the diamond surface and its research progress in mechanical properties from the perspective of plating materials. The application scope and advantages of plating technology to plating materials are expounded. The design idea of metal coating on diamond surface with low interface stress, high bonding force and suitable strength by optimizing plating technology and cross-using plating technology is expounded. Then future research trend of diamond surface metallization is proposed.

1. Introduction

Due to its high hardness and excellent wear resistance, diamond is the most suitable abrasive for manufacturing tools of hard material processing. It has been widely used for manufacturing tools of cutting, grinding, polishing and dressing. However, due to the small size of the diamond particles, diamond tools must be embedded or bonded together with metal, ceramic or resin and other binders in order to achieve efficient processing of the workpiece. However, diamond bonded by covalent bond has high interfacial energy, which makes it difficult for the bond to wet effectively on the surface of diamond, and the interfacial bonding force is poor, resulting in the bond holding force on diamond is small. When the cutting force is applied, diamond is easy to fall off prematurely, which reduces the service life and processing efficiency of diamond tools.

At present, surface modification of diamond is generally used at home and abroad to lower the interfacial energy between diamond and binder, and improve the bonding force between diamond and binder\textsuperscript{[1–5]}. The method of metallization of diamond surface is to uniformly coat a layer of metal film on diamond surface by means of surface treatment technology. Metal film makes diamond possess metallic properties and enhances the chemical affinity of metallized diamond with metals and their alloys. The thickness of the coating can range from nanometer to millimeter\textsuperscript{[6–9]}. The coating category can be divided into single metal coatings, alloy coatings and composite coatings, which can be mechanically embedded or chemically bonded with diamond surface\textsuperscript{[10–12]}.
This paper reviews the important research reports on diamond surface metallization at home and abroad in the past ten years from the perspective of plating materials, and points out the existing problems in the research and the development direction of future research, in order to provide technical support and reference information for basic research and engineering applications in related fields.

2. Single metal coating

Single metal coating means that the coating contains only one kind of metal or the metal carbide. The commonly used metal elements can be divided into three categories according to their interaction with diamond [3, 13]: (1) non-carbide forming elements, such as Cu, Zn, Sn, do not react with diamond and do not wet diamond; (2) weak carbide forming elements, such as Fe, Co, Ni, etc, are also known as graphitized elements. (3) Strong carbide forming elements, such as Ti, Cr, W, Mo, V, Nb, can react with C on the surface of diamond to form carbides and epitaxially grow on the surface of diamond.

2.1. Non/weak carbide forming element

For non/weak carbide forming elements, Ni and Cu are the main substrates for diamond coatings, and plating methods are mainly electroless plating and electroplating [14–16].

2.1.1. Electroless plating and electroplating

As diamond is an insulator, it is necessary to electrolessly plate a conductive film before electroplating, and then thicken the plating by electroplating or continue plating other metal plating. Therefore, electroless plating and electroplating are often used in combination to achieve large-scale production [17].

Based on the strict requirements of electroless plating and electroplating on the technological properties, in recent years, a large number of scholars have conducted in-depth and systematic studies on the influence of pretreatment of electroless plating and electroplating, plating composition, plating process and post-plating treatment on the coating properties.

2.1.1.1. Pretreatment of plating

As diamond itself has no catalytic effect on metal deposition, the pretreatment process of surface degreasing, sensitization and activation of diamond surface should be carried out before electroless plating and electroplating. Technological process is shown in figure 1 [10, 11, 18–23].

The commonly used technological formulation and operating conditions are as follows [18, 23–26]:

Degreasing: 5~10 g l⁻¹ NaOH, boil for 10~30 min, rinse to neutral with distilled water. Its purpose is to remove oil stains from the diamond surface.

Roughening: 10% nitric acid solution or mixed acid (concentrated nitric acid + concentrated sulfuric acid), boil for 20~30 min, rinse to neutral until distilled water. Its purpose is to roughen the surface of the diamond and increase the hydrophilicity of the diamond.

Sensitization: 10~20 g l⁻¹ SnCl₂·2H₂O, 40~48 ml l⁻¹ HCl, operating temperature is room temperature for 8~10 min. Its purpose is to adsorb some reducing agent on the surface of the diamond.

Activation: PdCl₂ is 0.2~0.5 g l⁻¹, HCl is 10 ml l⁻¹, operating temperature is room temperature for 10 min. Its purpose is to obtain a reactive center.

Dispergation: 5 g/L NaOH, its purpose is to dissolve Sn²⁺ or Sn⁴⁺ to expose the metal palladium which acts as a catalytic activity during electroless plating.

Sensitization and activation are the most critical steps in the pretreatment of diamond plating, which determine the strength of the combination between the coating and the diamond, as well as the continuity of the metal coating [19, 27]. In order to reduce the pretreatment sensitization and activation process, Hu et al [27]...
improved the strong acid colloidal palladium and developed a salt-based colloidal palladium solution by using the method of salt instead of acid. Huang [28] used this technology to electrolessly plate nickel on the diamond surface with particle size of 10 μm, and obtained the optimum electroless plating process of diamond micropowder. Chen et al [29] used this technology to prepare electroplated diamond tools. Results show that there is no obvious boundary between diamond and deposit, and there is nickel-cobalt junction at the interface, which improves the bonding strength between coating metal and diamond. Literature [30–32] considered that the hydrophilicity of micron diamond surface could be improved by immersing diamond in chromic acid pickling solution before plating.

2.1.1.2. Components of plating solution

The electroless plating bath mainly consists of metal salts (nickel sulfate, nickel chloride), reducing agents (sodium hypophosphate, formaldehyde, borohydride), complexing agents (sodium succinate), stabilizers (thiourea), and pH regulators (NaOH, HCl, acidic plating solution with pH 4–6, alkaline plating solution with pH 8–10). The electroplating bath mainly consists of main salts (nickel sulfate, nickel sulfamate), anode activator (nickel chloride, sodium chloride), buffer (boric acid) and wetting agent (sodium dodecyl sulfate). The concentration of components has a significant influence on the stability of the plating solution and the rate of weight gain of the coating, which determines the quality of the coating.

Liu et al [3] studied the influence of the concentration of NiSO₄ and NaH₂PO₂ on the coating quality, and the results showed that increasing the concentration of NiSO₄ was beneficial to improve the deposition rate, but the concentration was not too high, otherwise it would lead to too high concentration of Ni²⁺ in the solution, resulting in the production of nickel phosphate, and the stability of the plating solution would be reduced, and it is suggested that the concentration of NiSO₄ and NaH₂PO₂ was 3–5 g/l. Fang et al [33] found that NaCl was the main factor affecting the conductivity of electroplating solution. When the mass concentration was 5–30 g l⁻¹, the impact toughness of coated diamond increased with the increase of its concentration. When the mass concentration of NiSO₄ is between 100–250 g l⁻¹, the impact toughness of coated diamond increases with the increase of the concentration. Simakin et al [34] found that the deposition rate was not only affected by the concentration of main salts and reductants, but also by their concentration ratio. When the molar concentration ratio of Ni²⁺/H₂PO₂ was 0.25–0.6, the deposition rate was higher and reached the highest value when the molar concentration ratio was 0.3–0.45 (bath pH = 4.6).

2.1.1.3. Plating process

Electroless plating process parameters mainly include plating solution concentration, temperature, stirring rate, etc; electroplating process parameters mainly include cathode current density, pH value, and rotational speed, etc. These parameters have an impact on the quality of the diamond coating. Ren et al [23] used acid electroless nickel plating solution to electroless nickel plating on the surface of diamond powder. It was found that the plating temperature, time and pH of the plating solution had significant effects on the stability of the plating solution, deposition rate and coating quality. The diamond surface can not be well coated if the plating time is too short. On the contrary, the plating solution is unstable and easy to decompose if the plating time is too long. Fang et al [35] showed that when the electroplating current is 2–6 A, the nickel plating rate increases with the increase of current, the coating is more uniform and dense, the leakage plating phenomenon is reduced, and the impact toughness and compressive strength of nickel plated diamond is increased accordingly.

In order to improve the binding force between general coating and diamond, researchers have carried out a large number of experiments and researches to improve and innovate new processes of electroless plating and electroplating, such as the method of plating ‘spiny’ nickel coating, ultrasonic plating, etc. Studies have shown that [17, 21], compared with diamond smooth coating, the rough surface of the diamond coating is more conducive to improve the bonding force between nickel-plated diamond and resin, ceramic and other bonding agents, and the growth morphology and quantity of nickel spines mainly depend on the nickel plating process. Fang et al [36] studied the effect of plating current and time on the properties of resin-based nickel-plated diamond grinding wheel. It was considered that the nickel-plated diamond layer prepared by high current and short time plating process had a high density, slow growth of nickel spines, high bonding strength between the coating and resin, and the wear degree of the grinding wheel decreased by 29.43%. Under the same plating process and weight gain rate, pretreatment of diamond titanium plating and then nickel plating, coating nickel nodules more round, and the coating and diamond produced chemical bond, which enhanced the resin’s holding force on diamond.

Although the spiny-shapped nickel plating layer enhances the bonding force between the plated diamond and the matrix to a certain extent, as Ni is a high-toughness metal, an excessively thick plating layer inevitably affects the blade edge. To this end, Luo et al [37] studied the effect of weight gain rate on the performance of Ni-plated diamond tools. When the weight gain rate was 30%, the surface shape of the diamond wire saw obtained by nickel-diamond composite plating was the best, the deposition density of diamond particles was the highest, and
the cutting performance was the best. Yang [20] used the pulse barrel plating process to increase the diamond’s compressive strength by 39% when the diamond gained weight by 50%.

2.1.1.4. Post-plating treatment
In order to eliminate the interfacial stress between diamond and Ni-coated diamond, Fang et al [38] studied the post-plating heat treatment of Ni-coated diamond. Results showed that the average compressive strength of Ni-coated diamond increased slightly compared with that of non-heat-treated Ni-coated diamond, and did not affect the surface morphology of the coating, as shown in figure 2.

2.1.2. Other plating methods
In addition to electroless plating and electroplating, in the literature [39, 40], the surface of the diamond was plated with Ni by magnetron sputtering. It was found that, compared with conventional electroless plating, magnetron sputtering could obtain uniform and dense metal coating on the surface of diamond, which has good adhesion to the substrate. Zhang et al [41] obtained uniform metal coating by sputtering nickel on Diamond micro-particles by means of rocking and vibration, and prepared electroplated diamond wire saw with plated diamond. The results show that the improved device can effectively disperse the fine diamond powder, achieve uniform nickel plating on the surface of diamond, strengthens the combination of diamond and coating, and significantly reduce the amount of diamond falling off, as shown in figure 3.

2.2. Carbide forming elements
The structural condition of the chemical combination of diamond and binder is that the strong carbide forming elements, such as Ti, Cr, Mo, etc, in the coating epitaxially grown on the surface of diamond into a uniform, continuous and thin carbide layer [1, 3, 42–44]. According to whether the interface reaction occurs during the plating process, the plating methods of the carbide forming elements are classified into two types: (1) plating methods without interface reaction; and (2) plating methods with interface reaction.

2.2.1. Non-interfacial reaction plating methods
Non-interfacial reaction plating methods often adopt the method of magnetron sputtering. In order to form a chemical bond between the coating and the diamond, the coated diamond needs high temperature heat treatment (>800 °C) to cause interfacial reaction to form carbides [42].

Liu et al [45] studied the interfacial bonding state of magnetron sputtered Ti and Mo diamond in Fe-based, Ni-based and Co-based binders. The results show that the bonding strength between the coated diamond and the binder is related to the hot pressing process. When the hot pressing temperature is 830 °C~840 °C, only local
chemical bonding occurs between coatings and diamond, while most of the coatings can not react with diamond to form carbides, but melt or diffuse into the binder. Li et al. [46] studied the interface bonding state between titanium-coated diamond and matrix by magnetron sputtering. The results showed that it was difficult to form strong interface bonding between titanium-coated layer and diamond when the hot pressing temperature was 700 °C~840 °C. Zhu et al. [8] used DC magnetron sputtering to deposit Cr on the surface of diamond, and the post-plated diamond was annealed at 300~600 °C under ultra-high vacuum (5 × 10⁻³ Pa). Interfacial diffusion and reaction behavior between Cr film and diamond were studied. It was found that the Cr film and diamond had strong interfacial diffusion. Cr element could diffuse to the diamond surface and form a carbide layer of Cr at the interface. The interfacial diffusion reaction of Cr/diamond is controlled by the diffusion process of carbon. Increasing the heat treatment temperature or prolonging the heat treatment time can promote the diffusion reaction, and interfacial reaction products change from Cr₂C₃ to Cr₇C₃.

2.2.2. Interfacial reaction plating method
The plating methods with interfacial reaction mainly include salt bath plating, chemical vapor deposition plating, high temperature vacuum diffusion plating and vacuum micro-evaporation plating.

2.2.2.1. Salt bath plating
Salt bath plating is to put diamond into chloride salt bath mixed with metal powders such as Ti, Cr, W, etc, and then put it into high temperature furnace for salt bath treatment at 850~1000 °C for 1~2 h. Under the action of high adhesion of molten salt and capillary force, carbide coating is formed on the surface of diamond. A large number of studies have been carried out on salt bath-plated diamond at home and abroad. Technological process is as follows [42, 47–50]: diamond purification, activation → uniform mixing of metal salts (BaCl₂, NaCl, KCl, Ti₆C₄₀, deoxidizer) → add metal powder (Ti, Cr) and pre-treated diamond into the mixed molten salt → put the mixture into the crucible → high temperature sintering (>800 °C, argon or air atmosphere) → Boiled to remove slag and then filtered → Coated diamond.

Composition, phase structure and properties of salt bath titanium plating on diamond surface have been studied [47, 50, 51]. It is found that after high temperature (950 °C) salt bath plating, a dense Ti-plated layer is formed on the diamond surface, and TiC is formed between the coating and the diamond, which enhances the bonding force between the coating and the diamond and improves the compressive strength, oxidation resistance and thermal corrosion resistance of the diamond.

Liang et al. [9] studied the Ti-coated diamond surface plated by molten salt under microwave irradiation. The results show that for the 170/200 mesh diamond, a dense Ti coating is formed on the diamond surface at 600 °C~700 °C, but no carbide is formed, as shown in figure 4(a); TiC appears in the coating at 800 °C~1000 °C, and most TiC grains size are about 0.8 μm, as shown in figure 4(b); the coating is completely TiC layer at 1100 °C, and the grain size is about 1.5 μm, as shown in figure 4(c); For diamonds with a particle size of 5 μm, a large amount of TiC nano-layered structure and shrimp bone-like structure containing a trace amount of CI are formed in the product heat-treated at 1100 °C, as shown in figure 4(d).

In addition to Ti plating in salt bath, Tang et al. [42] treated diamond with tungsten plating in high temperature salt bath at 900 °C~1000 °C. After W plating, the diamond still maintains a complete crystal shape with no melting corrosion on the crystal surface. The surface structure of the plated diamond is diamond (inner layer) + WC + W (outer layer), which significantly improves the bonding performance between the plated diamond and the matrix, and gives full play to the wear resistance of the diamond. Kong [52] found that the salt bath chromium plating on the diamond surface formed a uniform chromium carbide between the diamond and the coating, which enhanced the performance of the diamond after plating. Xiang et al. [49] formed a uniform and dense plating layer on the surface of diamond after plating Cr in salt bath at 850 °C. The carbides of the plating layer are Cr₃C₂ and Cr₇C₃. The compressive strength of Cr-plated diamond is increased by 25.7% on average.

2.2.2.2. Chemical vapor deposition (CVD)
Chemical vapor deposition is a process in which a metal-plated gaseous compound (such as a halide) is introduced into the reaction chamber with diamond under certain temperature, pressure and time conditions, so that the gaseous materials will undergo thermal decomposition or chemical reaction with diamond to form a coating [2, 4, 53–56].

Wang et al. [33] plates titanium on the surface of diamond by CVD method, and TiC is generated on the surface of diamond, improving the performance of diamond and Ti-coated diamond tools. Wang et al. [55] used H₂ and WF₆ as reactive gases to deposit tungsten on diamond surface by CVD. The results show that uniform and dense coatings with nanometer thickness could be obtained on diamond powder surface by CVD at 580 °C~685 °C, but no carbide is found. The optimum process for tungsten plating by CVD method is as follows: 670 °C, 2 min, 1 L min⁻¹ of H₂ flow rate and 2 g min⁻¹ of WF₆ consumption. Compared to the
unplated diamond segments, the flexural strength of the diamond segments is increased by nearly 40%. Vesna et al. [56] plated chromium on the surface of diamond with gaseous chromium iodide as reaction gas by CVD method. Chromium plating containing Cr$_7$C$_3$ and Cr$_3$C$_2$ is obtained at the temperature of 700 °C–1000 °C.

2.2.2.3. Vacuum thermal diffusion plating

Vacuum hot diffusion plating can be carried out without special plating equipment. The process is as follows [57, 58]: weighing a certain proportion of diamond and metal powder → uniformly mix diamond, metal-plated powder, wetting agent and pore forming agent → evenly loading mixture into crucible (molybdenum crucible for Ti and graphite crucible for B) → placing in vacuum furnace → high temperature heat treatment → cooling with furnace to room temperature.

Luo et al. [57] used this method to coat the surface of diamond in a common vacuum furnace. It is studied that the effects of plating temperature and holding time on the surface morphology, thickness and phase of the diamond surface coating. The oxidation resistance of titanium-coated diamond and its binding state in iron matrix were analyzed. The results show that continuous coating can be formed on the surface of diamond at lower temperature. The suitable coating temperature is 720 °C–780 °C. The ideal coating thickness can be obtained by adjusting the coating temperature and holding time, as shown in figure 5. The coating improves the high temperature thermal invasion resistance of diamond, makes the interface between titanium-coated diamond and matrix better, and improves the flexural strength of composite matrix.

Bai et al. [58] used high-temperature vacuum evaporation to deposit boron on the surface of micron-sized diamond. And then diamond–aluminium matrix composites were prepared by vacuum hot pressing sintering of the plated diamond and Al powder. The results show that the surface of the diamond is coated with a thickness of 1 μm. The coating is made of boron and boron carbide. It avoids the direct contact reaction between diamond and aluminium in the preparation of boron-plated diamond/Al-based composites to form Al$_4$C$_3$ hydrolysis phase.Selective bonding between non-plated diamond and aluminium matrix is eliminated. AlB$_2$ is formed at the coating/Al interface, which achieves chemical bonding between diamond and metal matrix.

Wei et al. [59] used high-temperature vacuum evaporation to coat tungsten on the surface of diamond. It is found that tungsten plating is a process of atomic diffusion reaction. The phase of the coating is affected by the plating temperature and time. W$_2$C is formed in the coating at the plating temperature of 950 °C. WC phase appears in the coating along with the W phase and the W$_2$C phase at the temperature of 1000 °C. Abyzov et al. [60] obtained Tungsten Coatings with different thickness on diamond surface by high-temperature hot diffusion plating. The results show that the thermal conductivity can reach 907 W m$^{-1}$·K$^{-1}$ when the thickness
of the coating is 110 nm. Bai [61] and Zhang [62] used thermal diffusion method to coat tungsten on diamond surface. A series of studies were carried out on the coating process, coating thickness and phase evolution. The thermal conductivity of the optimized coatings could reach 721 W m\(^{-1}\) K\(^{-1}\).

2.2.2.4. Vacuum micro-evaporation plating
In order to solve the problems existing in the above plating methods, such as less single plating amount, higher plating cost, higher plating temperature and thermal damage to diamond, Yanshan University developed vacuum micro-evaporation plating technology for superhard materials [3], and achieved many leading technological achievements at home and abroad. Vacuum micro-evaporation plating is a process in which the forming elements of strong carbides become more active at a lower temperature, obtain energy and increase the amplitude, and react with diamond to form carbides and form a coating by utilizing the principle of the reduction of metal vapor pressure under the vacuum state. This method is favored by many researchers because of its advantages of large single plating, high plating quality and low cost [63–65].

Wang et al [53, 63, 66] used vacuum micro-evaporation plating to coat Ti, Mo, W and Cr on the surface of diamond to improve the bonding force between diamond and binder. The results show that TiC layer is formed by interfacial reaction between titanium plating layer and diamond, which plays a protective role on diamond during sintering process and significantly improve the service life and efficiency of diamond tools. Wang et al [1, 67] shows that the formation of carbides is related to the diffusion of atoms between coatings and diamonds when vacuum micro-evaporation titanium plating on diamond surface. TiC is formed close to the surface of diamond with high C concentration, and pure Ti layer is gradually formed far away from the diamond coating due to insufficient C atom supply. When the coating structure is diamond \( \rightarrow \) TiC \( \rightarrow \) Ti at the plating temperature of 750 °C, which improves the strength and impact strength of coated diamond, and enhance bonding force with the binder (\(\geq 140\) MPa); However, the surface properties of diamond are degraded when the plating temperature exceeds 800 °C.

![Figure 5. Titanium layer thickness maintained at different plating temperatures for 90 min. (a) 720 °C; (b) 740 °C. (c) 780 °C. (d) 820 °C [57].](image)
Zhang et al [68] found that there was no carbide formation in the coating at the temperature of 800 °C, the coating is discontinuous, and there are defects such as holes and pits; A small amount of W₂C appears in the coating at 900 °C for 1 h, and the integrity of the coating is better than 800 °C, but there is a small amount of uncovered area at the junction of the crystal plane, as shown in figure 6(a); WC phase is formed in the coating at 900 °C for 2 h, the coating has become complete and dense, and there are no defects on the diamond surface, as shown in figure 6(b); With the further prolonging time, the phase composition of the coating is basically constant, the coating is thicker, W particles grow up, bond or even stack. In addition, unlike literature [1, 60], there is not any graphitization on diamond surface in literature [61] under experimental conditions.

2.2.2.5. Other new plating methods
Based on the factors of chemical co-deposition, magnetron sputtering without chemical bonding, less single CVD plating, high temperature of salt bath plating, difficult treatment after plating, and special equipment for vacuum micro-evaporation plating.

Gu [69] used microwave heating method to plating Ti on diamond surface. The plating process was as follows: diamond purification, activation treatment → uniform mixing of diamond and TiH₄ (mass concentration 1:0.6) → placing in ceramic crucible → microwave heating treatment (660 °C~860 °C, 1 h, argon protection) → cooling with furnace to room temperature → distilled water cleaning → vacuum drying (75 °C, 4 h). The post-plating performance test shows that when the plating temperature is 710 °C~810 °C, a uniform continuous titanium coating layer is formed on the diamond surface, and TiC is formed. Increasing the plating temperature can increase the thickness of the TiC layer, but also coarsen the coating grains. When the plating temperature is 760 °C, the bonding force between diamond and Ti-coated layer is the greatest. A new method of coating tungsten metal film on diamond surface is proposed by Xiang et al [22]. This method utilizes the volatile characteristics of W oxides in water vapor to reduce W oxides containing catalysts by H₂/H₂O mixed gas at 750~900 °C. It can coat tungsten metal on diamond surface with a coverage of more than 90%. The results show that the phases of the coating are W and W₂C at 750 °C, while phases of the coating are WC and W₂C at 900 °C for WC being formed by further reaction of W with C on the diamond surface.

2.2.3. Carbide formation mechanism
The carbide formation process is a typical diffusion reaction process. The strong carbide forming elements and the C on the diamond surface have mutual reaction and diffusion behavior at a certain plating temperature and pressure.

During the interfacial reaction process, the plated metal with a larger atomic radius is difficult to diffuse into the diamond lattice, while the C with a small atomic radius diffuses unilaterally into the coated metal. Due to the gradient of diffusion concentration, carbides are formed on the diamond surface, low carbon compounds are formed near the coating side, and pure metal coating is formed where C can not migrate in the coating. Based on the above diffusion theory, literatures [3, 19, 44] propose a crystal model of diamond surface metallization, which consists of three layers of materials, including carbide layer, alloying layer and pure metal plating layer from the inside to the outside, as shown in figure 7. The thickness of the carbide layer is generally several hundred nanometers, which plays a key role in the metallization of the diamond surface; the thickness of the alloyed layer is generally several micrometers, which has good bonding to carbide layer; the pure metal coating can alleviate the difference of physical and chemical properties between diamond and matrix, and reduce internal stress.

The formation of carbides at the interface between diamond and binder can not be at the expense of thermal damage on the surface of diamond. Thermodynamic data of the reaction of Ti, Cr, W and their oxides with diamond and O₂ are calculated and analyzed in [21, 47, 49, 70, 71], as shown in table 1. For the thermodynamic data in table 1 are obtained under standard conditions, it is pointed out in [49, 72] that the formation temperature of carbides should be about 150 °C higher than the minimum temperature calculated by
thermodynamics. It can be seen from table 1 that the binding ability of Ti, Cr and W elements to O₂ is greater than that of diamond. Therefore, according to literatures [49, 72], the interaction between strong carbide forming elements such as Ti, Cr, W and diamond does not occur directly, but by means of the strong affinity of Ti, Cr, W and O₂. Firstly, elements, such as Ti, Cr and W, capture the chemisorbed oxygen on the diamond surface to protect the carbon on the diamond surface from converting to graphite. Then, at high temperature, the reductive reaction of Ti, Cr, W oxides and C on the diamond surface produces a layer of carbides evenly distributed on the diamond surface. A comparison of the plating methods of the carbide forming elements on the diamond surface is shown in table 2.

3. Alloy Coatings

With the rapid development of modern industry, traditional and single metal-coated diamond tools can no longer meet the need of processing performance. Instead, it is to improve the coating process and improve the performance of diamond surface coating. Compared with vacuum plating and salt bath plating, electroless plating and electroplating are often used for alloy coating. By adding different metal salts as reducing agents in electroless plating and electroplating bath, and adopting appropriate process, two or more metal elements can be co-deposited, such as Ni–P, Ni–B, Ni–W–P, etc.

3.1. Alloy coating without carbide forming elements

The alloy coatings without carbide forming elements are mainly Ni–P coatings for diamond plating, and the research content is bath composition, plating process and the phases of coating.

3.1.1. Bath composition and plating process

Mao [24] studied the composition and process of electroless Ni–P alloy plating on the diamond surface. The results showed that increasing plating temperature could increase plating rate, but too fast plating rate would lead to the decrease of phosphorus content in the coating, the increase of stress and porosity of the coating, and the decrease of corrosion resistance of the coating. The surface of Ni–P coated diamond was graphitized after high temperature treatment at 800°C. However, compared with the non-plated diamond, the high temperature thermal corrosion resistance of the coated diamond was improved. Yao et al [73] studied the effect of pH on the morphology of electroless Ni–P coating on the surface of diamond micropowder. The results show that when the pH of the plating solution is less than 13, a complete coating cannot be formed; when the pH is 13, a uniform, dense and complete coating is formed; when the pH is greater than 13, the coating will fall off.

3.1.2. Phases and function of coating

Guan et al [25] used electroless plating, electroless plating + electroplating to coat Ni–P alloy on the surface of diamond respectively, And studied the properties of the coated diamond. The results show that the Ni–P alloy coating is amorphous. Both methods can improve the compressive strength and thermal corrosion resistance of diamond coated, but the latter is better. The amorphous Ni–P alloys were transformed into Ni₃P, Ni₅P₂ and Ni after crystallization at 400°C. The compressive strength of diamond coated after crystallization was slightly lower.
| Chemical reaction equation | $\Delta G_T = A + BT$ kJ mol$^{-1}$ | $\Delta G_T (800 \degree - 900 \degree C)$ | Chemical reaction equation | $\Delta G_T = A + BT$ kJ mol$^{-1}$ | $T\Delta G_T = 0$ K |
|-----------------------------|---------------------------------|--------------------------------|-----------------------------|---------------------------------|-------------------|
| Ti + O$_2$ = TiO$_2$        | $\Delta G_{T1} = -943.84 + 0.184$ T | $\Delta G_{T1} < \Delta G_{T2} < 0$ | TiO$_2$ + 3 C(d) = TiC + 2CO | $\Delta G_T = 533.41 - 0.48$ T | 1111.3 (838 $\degree$C) |
| Ti + C(d) = TiC             | $\Delta G_{T2} = -183.1 + 0.01008$ T |                                    | 1.5 Cr$_2$O$_3$ + 6.5C(d) = Cr$_3$C$_2$ + 4.5CO | $\Delta G_T = 1089.5 - 1.102$ T | 988.66 (715 $\degree$C) |
| 2Cr + 1.5O$_2$ = Cr$_2$O$_3$ | $\Delta G_{T3} = -1128.6 + 0.266$ T |                                    | 1.5 Cr$_2$O$_3$ + 8.5C(d) = Cr$_3$C$_3$ + 5.5CO | $\Delta G_T = 2584.14 - 2.672$ T | 967.12 (694 $\degree$C) |
| 3Cr + 2 C(d) = Cr$_3$C$_2$  | $\Delta G_{T4} = -791 - 0.0177$ T | $\Delta G_{T3} < \Delta G_{T4} < 0$ |                                            |                                    |                   |
| 7Cr + 3 C(d) = Cr$_7$C$_3$  | $\Delta G_{T5} = -153.6 - 0.0372$ T |                                    |                                            |                                    |                   |
| W + 1.5O$_2$ = WO$_3$       | $\Delta G_{T6} = -833.45 + 0.24$ T | $\Delta G_{T5} < \Delta G_{T7} < 0$ | WO$_3$ + 4 C = WC + 3CO | $\Delta G_T = 442.17 - 0.5$ T | 884.34 (610 $\degree$C) |
| W + C(d) = WC               | $\Delta G_{T7} = -37.66 + 0.0017$ T |                                    |                                            |                                    |                   |
Table 2. Typical plating method for carbide forming elements on diamond surface.

| Technical characteristics | Magnetron sputtering method | Salt bath plating | Chemical vapor deposition | High temperature vacuum thermal diffusion plating | Vacuum micro-evaporation plating |
|---------------------------|-------------------------------|-------------------|---------------------------|-----------------------------------------------|----------------------------------|
| Coating composition       | Ti, Cr, Mo and corresponding carbides | Ti, Cr, Mo, W and corresponding carbides | Ti, Cr, Mo, W and corresponding carbides | Ti, Cr, W and corresponding carbides | Ti, Cr, Mo, W and corresponding carbides |
| Plating structure         | The coating is physically attached to the diamond surface | Carbide formation | Point Epitaxy Growth of Carbide + Surface Metal Layer | Carbide formation | Complete Epitaxial Growth of Carbide + Surface Metal Layer |
| Binding state with the matrix | Physical bonding, Low bonding strength | Metallurgical bonding | Metallurgical bonding | Metallurgical bonding | Metallurgical bonding, High bonding strength |
| Coating temperature       | ≤400 °C                       | ≥850 °C           | ≥850 °C                   | ≥800 °C                                      | 650 °C–750 °C                       |
| Thermal damage of diamond | No thermal damage             | Severe thermal damage | Severe thermal damage     | Thermal damage                               | No thermal damage                  |
| Single plating capability | Low                           | High              | Low                       | High                                         | High                              |
than that of non-crystallization. Liu et al [6] studied the electroless plating of high-phosphorus Ni–P alloy on diamond surface, and discussed the phase composition and growth mode of Ni–P coating. The results show that a complete, uniform and dense Ni–P alloy coating is formed on the surface of diamond with a thickness of 4–6 micron. Ni and P coexist in the alloy coating, which are uniformly distributed and amorphous. The contents of Ni and P are 86.81 wt% and 13.19 wt%, respectively. The coatings are transformed into crystalline Ni₃P compounds after heat treatment, and the Ni–P coatings first nucleate at the edges and corners of diamond, and then grow in lamellar or convex spheres. There is a clear interface between the coating and diamond, although it is well combined.

3.2. Alloy coating containing strong carbide forming elements
Since Ni–P plating is only deposited on the diamond surface without any chemical bonding, there is no significant improvement in the bonding force between the diamond and the binder. Therefore, the ternary and quaternary alloy coatings are prepared by adding other metal salts as reducing agents in Ni–P and Ni–B electroless plating solution.

Xiang et al [74] used electroless plating to plate Ni–W–P coating on the surface of diamond. The results show that the diamond is completely coated by the plating. The coating is composed of cellular particles with uneven size and irregular shape. The contents of Ni, W and P are 75.20%, 13.57% and 4.83% respectively which is analyzed by EPMA. After heat treatment at 850 °C in the air, the surface of the coating becomes uneven, forming a honeycomb structure with more voids. Though compressive strength of the plated diamond is reduced by heat treatment, the compressive strength and thermal corrosion resistance of the plated diamond are greatly improved comparing with the non-plated diamond. Hu et al [75] find that the Ni–W–P coating is amorphous and there is no carbide on the surface of diamond when electroless plating Ni–W–P coating on the surface of diamond. However, after heat treatment at 800 °C and above, WC is formed on the surface of diamond coated. Duan et al [72] electroless plated Ni–W–B alloy on the surface of diamond and analyzed the thermal damage characteristics of Ni–W–B diamond plated at high temperature. The results show that the Ni–W–B plated diamond is mainly surface damage at high temperature, while the un-plated diamond is mainly caused by body damage. Han [76] electroless plated Ni-Mo-P alloy on the surface of diamond, and obtains high hardness and heat corrosion resistance coatings.

4. Composite coatings
There are two kinds of composite coatings: one is a layered coating made of two or more layers of metal, such as adding Cr layer to the Ni layer, and the plating method is a composite plating method such as electroless plating + electroplating, electroless plating + vacuum plating; the other is a coating formed by the uniform dispersion of solid particles in the metal, also known as the dispersion coating. The method is to add an appropriate amount of insoluble metal particles to the plating solution for plating, so that the particles are uniformly co-deposited in the metal plating layer [76–78].

Duan et al [21] used vacuum plating to coat Ti–Cr composite coating on the surface of diamond. There are TiC, Cr₇C₃, Cr₇C₃ carbide formed on the diamond surface, and C of the carbide comes from the diamond itself. The combination of the coating and the diamond is good, and there is no crack and shedding phenomenon, and the chemical bonding between Ti–Cr plated diamond and binder is achieved. Zhong [79] coated Ti–Ni composite coating on diamond surface by vacuum micro-evaporation plating and electroplating respectively, and studied the brazing performance of Ti–Ni diamond. The results show that Ti–Ni coating improves the brazing performance of diamond. On the one hand, Ti coating isolates the direct contact between Ni and diamond, thus avoiding the graphitization of diamond in brazing, on the other hand, Ni coating prevents and reduces the oxidation tendency of Ti and diamond. The bonding strength of brazing interface between Ti–Ni plated diamond and matrix is greater than 140 MPa. Hu et al [27] first coated Ni–W–P alloy on the surface of diamond by electroless plating, and then electrolessly plated Ni–Ti–RE on the surface of Ni–W–P coated diamond by adding appropriate amount of titanium metal powder to the plating solution. It is found that this method realizes the deposition of granular metal Ti on the diamond surface. TiC is formed on the diamond surface after heat treatment at 900 °C, which significantly enhances the bonding strength between diamond and matrix metal.

5. Summary and prospect

(1) The surface of the diamond is metallized by electroless plating or electroplating technology to have metal characteristics, make up for surface defects, improve compressive strength and high temperature corrosion resistance, and enhance the bonding force between diamond and resin bond. However, the coating does not
form a chemical bond with the diamond and has interfacial stress, which is not suitable for metal and ceramic bond tools.

(2) Using salt bath plating, chemical vapor deposition, high temperature vacuum diffusion plating, vacuum micro-evaporation plating and other technologies, carbide forming elements such as Ti, W, Cr, B were coated on the surface of diamond to achieve chemical bonding between diamond and binder, thus significantly improving the holding force of binder to diamond. The shortcomings are the high plating temperature of salt bath plating and vacuum diffusion plating, the small amount of single CVD plating, special equipment needed for vacuum micro-evaporation plating, etc, which have certain limitations in improving the performance of diamond tools.

(3) By adjusting bath composition or adding metal particles, as well as using composite plating technology, the alloy coating and composite coating on diamond surface are achieved, which can improve the performance of the coating and enhance the bonding force between binder and diamond. However, the technology is still at the stage of laboratory and theoretical research, and has not been applied in large-scale industrialization.

(4) It is an effective way to improve the performance of resin-based diamond tools by plating the ‘spiny-like’ coating on the diamond surface using Electroless plating and electroplating. In order to give full play to the role of ‘spiny-like’ coating, the growth mechanism, surface morphology reconstruction and constitutive equation of ‘spiny’ coating should be studied in depth.

(5) In order to achieve a breakthrough in the higher performance of diamond surface metallization, it is necessary to break the limitations of the existing plating technology, and utilize the advantages of various plating methods to innovatively utilize, combine and develop various composite plating methods. In addition, it is necessary to strengthen the exchange of metallization of diamond surface between academia and industry, reach a common understanding, form a unified guiding norm, formulate relevant industry standards, and achieve standardization.

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