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石墨烯改性金刚石刀具的 瞬时转化机理与应用基础研究

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Research on Instantaneous Transformation Mechanism and Fundamental Application of Graphene-Modified Diamond Cutting Tools

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摘 要

金刚石被称为保障航空航天、微机电系统、生物医学和核能等领域产品高性能的“极端功能材料”，其中，金刚石刀具已经应用于超精切削和 3C 产品加工，但是其在高载、与黑色金属接触等条件下，易石墨化和非晶化转变等问题严重制约了金刚石刀具的广泛应用。将金刚石与石墨烯结合以获得优异的协同性能，是解决金刚石上述“软肋”的有效途径之一，被国家自然科学基金委列为 10000 个制造科学难题之一。大量研究以“增”的方式将石墨烯涂覆在金刚石表面，或通过化学气相沉积方式在金刚石表面直接生长出石墨烯，但难以克服结合强度有限、金刚石表面弱化或制备环境苛刻等难题。本论文针对重点领域中大量特殊工况下减摩、抗机械化学磨损需求，提出激光-机械结合新途径，制备出新型石墨烯改性金刚石刀具，系统研究了转化机理、调控机制、特性表征、抗磨损与结构失效机制，开展了航空航天碳纤维复材和黑色金属难加工材料的切削试验，有效提高了金刚石刀具的切削性能。本论文力图丰富金刚石-石墨烯异质结合及性能协同的多学科交叉理论，实现金刚石表面石墨烯瞬时共价键合改性新技术，为破解新型难加工材料加工瓶颈难题，提供高性能金刚石刀具，并有望为金刚石、金刚石涂层、石墨烯在机械、电子、航空航天等领域开辟新的应用前景。论文主要研究内容如下：

(1) 金刚石-纳米石墨-石墨烯共价新结构 (Covalent structure of Diamond-Nano-graphite-Graphene, CDGG) 的瞬时转化生成机理：以“原位瞬时转化”新思想，提出激光-机械结合新方法，首先验证了纳秒激光诱导高取向性石墨的存在性，并结合物理计算明晰了激光诱导金刚石向高取向性石墨的瞬时转化机理和激光器对诱导高取向性石墨的适配性；研究了石墨化对后续解理的影响，并结合能量计算揭示了机械解理高取向性石墨成石墨烯的瞬时转化机理；首次在空气中形成了一种稳定独特的 CDGG 共价新结构；最后基于表征分析和物理计算，归纳了原位 CDGG 的典型结构特征。

(2) 典型金刚石材料表面原位 CDGG 的可控制备：分析了影响原位 CDGG 结构特征形成的主要影响因素，建立了相应的试验平台；开展了金刚石厚膜表面原位 CDGG 的调控机制研究，探明了主要影响因素对石墨化和机械解理的影响规律；针对金刚石涂层（薄膜）的结构特征，构建了激光诱导涂层石墨化的有限元数值模型，建立了涂层表面精准石墨化的调控机制，提出了原位 CDGG 结构特征的快速评估手段和定量指标；发明激光-机械结合的金刚石-石墨-石墨烯瞬时转化新方法，实现了金刚石厚膜、金刚石涂层表面原位 CDGG 的可控制备和图案化。

(3) 原位 CDGG 的力学特性分析：构建了原位 CDGG 的分子动力学压痕模型，揭示了原位 CDGG 表层的形变特性；在 1~9N 的常规载荷范围开展摩擦学试验，探明了原位 CDGG 面对

超硬材料和黑色金属的摩擦学行为。结果表明，原位 CDGG 显著降低摩擦系数、提高表面韧性和抗磨能力，体现出金刚石、石墨烯的综合特性。常规载荷下（ $<10\text{N}$ ），相对原始金刚石，在面对 Si_3N_4 超硬材料的干摩擦下，CDGG 的稳定摩擦系数降低 29%~60%；在面对黑色金属的干摩擦下，CDGG 的稳定摩擦系数降低 49%~59%，相对磨损率降低一个量级。

（4）原位 CDGG 的抗磨损与结构失效机制：追踪了摩擦界面的生成与失效的演化过程，明晰了常规工况下原位 CDGG 的磨损抑制与结构失效机制；探明了原位 CDGG 在复杂工况摩擦过程中的结构变化，并验证了其结构的独特性；建立了原位 CDGG 的分子动力学摩擦模型，阐明了其润滑与机械化学磨损抑制机制。理论和试验综合表明，CDGG 协同了金刚石、石墨和石墨烯的优良性能，呈现出优异的承载、润滑、抗机械化学磨损和持久等特性。

（5）原位 CDGG 刀具的典型应用基础研究：针对航空碳纤维复材加工难题与高效精密制造迫切需求，研制出航空复材加工专用金刚石刀具，试验分析了原位 CDGG 制备对其切削性能、使用寿命和经济性的影响，结果表明，CDGG 的制备使刀具的铣削力、磨损量分别降低 38%、24%；针对金刚石刀具加工黑色金属难题，对标准数控金刚石车刀片进行 CDGG 改性，试验了 CDGG 车刀对黑色金属材料的切削性能，表明 CDGG 有效提升了刀具抗机械化学磨损性能，并阐明了其减摩抗磨机制；最后基于 CDGG 的其他优良特性，开展了微动损伤抑制和电加热除冰应用基础研究，为后续发展拓宽了思路。

关键词：刀具，金刚石，石墨烯，共价结构，减摩抗磨

ABSTRACT

Diamond is known as an "extreme functional material" that ensures high performance in products for aerospace, microelectromechanical systems, biomedical, nuclear energy, and other fields. Diamond cutting tools, in particular, have been utilized in ultra-precision cutting and processing of 3C products. However, challenges such as graphitization and amorphization under high loads and contact with ferrous metals severely limit the widespread application of diamond tools. Combining diamond with graphene to achieve excellent synergistic performance is deemed an effective approach to address these weaknesses of diamond and has been recognized as one of the 10,000 manufacturing science challenges by the National Natural Science Foundation of China. Numerous studies have focused on "adding" graphene to or directly growing graphene on diamond surfaces through chemical vapor deposition. Yet, overcoming issues like limited bonding strength, surface weakening of diamond, or harsh preparation conditions remains challenging. This paper proposes a novel laser-mechanical hybrid approach to prepare graphene-modified diamond cutting tools to meet the demands for reducing friction and combating mechanochemical wear in critical scenarios. The research systematically investigates the transformation mechanism, control mechanisms, characterization of properties, wear resistance, and structural failure mechanisms. Cutting tests on aerospace carbon fiber composites and ferrous metal difficult-to-cut materials have effectively enhanced the cutting performance of diamond tools. This work seeks to enrich the multidisciplinary theory underlying diamond-graphene heterojunctions and their property synergy. It aims to achieve a novel technique for the instantaneous covalent bonding of graphene on diamond surfaces, providing high-performance diamond cutting tools to overcome bottlenecks in machining new difficult-to-cut materials. Furthermore, it is expected to open new application prospects for diamond, diamond coatings, and graphene in the fields of machinery, electronics, and aerospace. The main research contents are as follows:

(1) Instantaneous transformation mechanism of in-situ covalent structure of diamond-nano-graphite-graphene (CDGG) on diamond surface: Based on the novel concept of "in-situ instantaneous transformation", proposing a laser-mechanical hybrid approach. Initially, the existence of laser-induced highly oriented graphite is validated, and the instantaneous transformation mechanism of laser-induced diamond to highly oriented graphite is elucidated in conjunction with physical computations, along with assessing the adaptability of lasers for inducing highly oriented graphite. The impact of graphitization

on subsequent cleavage is investigated, and the instantaneous transformation mechanism of mechanically cleaving highly oriented graphite into graphene is revealed through energy calculations. A stable and unique CDGG is instantaneously formed for the first time in air. Finally, typical structural characteristics of CDGG are summarized based on characterization analysis and physical computations.

(2) Controlled preparation of in-situ CDGG on typical diamond material surfaces: Analyzing the main factors influencing the formation of in-situ CDGG structural features, this study establishes a corresponding experimental platform. Research on the regulation mechanism of in-situ CDGG on diamond thick film surfaces is conducted, uncovering the impact of major factors on graphitization and mechanical cleavage patterns. A numerical model for laser-induced coating graphitization is developed for typical diamond coatings (films), elucidating the control mechanism of surface graphitization in coatings. Means and indicators for rapid quantitative assessment of the structural characteristics of in-situ CDGG are proposed. By inventing a laser-mechanical combined method for the instantaneous transformation of diamond-graphite-graphene, controlled preparation and patterning of in-situ CDGG on diamond thick films and diamond coating surfaces are achieved.

(3) Analysis of the mechanical properties of in-situ CDGG: A molecular dynamics indentation model of in-situ CDGG is constructed to reveal surface deformation characteristics. Friction experiments are conducted within the range of 1~9 N conventional loads, investigating the friction behavior of CDGG against superhard materials and ferrous metals. Results demonstrate that in-situ CDGG significantly reduces friction coefficients, enhances surface toughness and wear resistance, showcasing the comprehensive properties of diamond and graphene. Under conventional loads (<10 N), in dry friction against Si_3N_4 superhard materials, the stable friction coefficient of CDGG decreases by 29% to 60% compared to pristine diamond. When facing dry friction against ferrous metals, the stable friction coefficient of CDGG decreases by 49% to 59%, with a reduction of one order of magnitude in relative wear rate.

(4) Wear resistance and structural failure mechanism of in-situ CDGG: The evolution process of friction interface formation and failure is tracked, elucidating the wear suppression and structural failure mechanisms of CDGG under typical conditions. Structural changes of in-situ CDGG during friction in complex conditions are explored, verifying the uniqueness of its structure. A molecular dynamics friction model of CDGG is established, elucidating its lubrication and mechanochemical wear suppression mechanisms. Theoretical and experimental results collectively demonstrate that the CDGG synergizes the excellent properties of diamond, graphite, and graphene, exhibiting outstanding characteristics in load-bearing capacity, lubrication, resistance to mechanochemical wear, and durability.

(5) Fundamental applications of in-situ CDGG tools: Addressing challenges in aerospace carbon fiber composite machining and the urgent need for efficient precision manufacturing, specialized diamond tools for aerospace composite machining are developed using CDGG. The impact of CDGG preparation on cutting performance, service life, and cost-effectiveness of these tools is tested and analyzed, showing the preparation of CDGG reduces milling force and tool wear by 38% and 24% respectively. To tackle issues in machining ferrous metals with diamond tools, standard CNC diamond turning inserts are modified with CDGG. Tests on the cutting performance of CDGG tools on ferrous metal materials demonstrate enhanced resistance to mechanochemical wear, elucidating their lubrication and anti-wear mechanisms. Finally, leveraging the other excellent characteristics of CDGG, fundamental research into applications for fretting damage suppression and electric heating de-icing is conducted, broadening the scope for future development.

Keywords: Cutting tool, Diamond, Graphene, Covalent structure, Lubrication and anti-wear