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Enhanced Mechanical Properties of Aluminum Oxide Coatings through the Incorporation of Carbon Nanotubes and Graphene Nanoplatelets: Insights from Recent Studies

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ABSTRACT

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This study presents a comprehensive review of the advancements in aluminum oxide (Al2O3) coatings, focusing on the incorporation of carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) to enhance their mechanical properties. The review systematically examines the fabrication techniques of these composite coatings, particularly utilizing the atmospheric plasma spray technique for their deposition onto low carbon steel substrates. The effects of CNT and GNP content on the densification, relative hardness, elastic modulus, and fracture toughness of the coatings are critically evaluated. The synergistic interaction between CNTs and GNPs leads to remarkable improvements in mechanical properties, with a significant increase in densification up to 97% of theoretical density. The addition of CNTs and GNPs results in a substantial enhancement in relative hardness by 52%, and elastic modulus by 48%. Most notably, the fracture toughness demonstrates an exceptional improvement of 200%, attributed to various toughening mechanisms including bridging, crack deflection, and interlocking provided by CNTs and GNPs. This systematic review provides valuable insights into the design and performance of advanced Al2O3 coatings reinforced with CNTs and GNPs, offering a promising avenue for the development of high-performance materials with enhanced mechanical characteristics.

Keywords: Aluminum oxide coatings, Carbon nanotubes, Graphene nanoplatelets, Mechanical properties, Synergistic reinforcement

INTRODUCTION

[†] Footnotes relating to the title and/or authors should appear here.

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In recent years, the exploration of advanced composite materials has gained substantial attention in various industries due to their potential to enhance the mechanical properties and functional performance of traditional materials. Aluminum oxide (Al2O3) coatings,

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renowned for their corrosion resistance and wear protection, have been extensively studied to further improve their mechanical characteristics. The integration of carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) into Al2O3 coatings presents a promising avenue to address existing limitations and to create hybrid materials with enhanced strength, hardness, and fracture toughness. While prior research has demonstrated significant improvements in the mechanical properties of Al2O3 composites reinforced with individual carbon-based nanomaterials, there remains a gap in the systematic analysis of the synergistic effects resulting from the combined inclusion of CNTs and GNPs. This study aims to bridge this gap by providing a comprehensive review and analysis of the advancements achieved through the incorporation of both CNTs and GNPs in Al2O3 coatings, shedding light on the underlying mechanisms and potential applications of these synergistically reinforced materials.

The current state of research in the field of advanced composite coatings highlights a significant focus on enhancing the mechanical properties of aluminum oxide (Al2O3) coatings through the incorporation of carbonbased nanomaterials. Carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) have emerged as promising reinforcements due to their exceptional mechanical characteristics and unique structural properties. Previous studies have individually explored the incorporation of CNTs or GNPs into Al2O3 coatings, revealing improvements in hardness, strength, and wear resistance [1].





However, a comprehensive investigation into the combined effects of CNTs and GNPs within Al2O3 coatings remains scarce. While existing research has demonstrated the potential of these nanomaterials to enhance specific properties, a systematic assessment of their synergistic influence on overall mechanical behavior, fracture toughness, and other critical parameters is lacking. This gap underscores the necessity of a comprehensive review and analysis that elucidates the combined effects of CNTs and GNPs on Al2O3 coatings, ultimately providing insights into the development of high-performance hybrid materials with broad applications in various industries [2].

The novelty of this research lies in the comprehensive exploration of the combined influence of carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) on the mechanical properties of aluminum oxide (Al2O3) coatings. While previous studies have individually investigated the impact of CNTs or GNPs, this research uniquely addresses their synergistic effects within the same Al2O3 matrix. The investigation aims to elucidate the mechanisms underlying the enhanced mechanical characteristics and improved fracture toughness observed in these composite coatings [3].



Figure 2. For catalytic asymmetric hydroformylation (AHF) of alkenes to chiral aldehydes, though a topic of high interest, the contemporary developments remain largely empirical owing to rather limited molecular insights on the origin of enantioselectivity.

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By systematically analyzing the interactions between CNTs and GNPs and their contributions to properties such as hardness, strength, and elastic modulus, this research contributes to a deeper understanding of nanomaterial-reinforced coatings. The ultimate goal is to provide valuable insights for the design and development of advanced materials with tailored mechanical properties, opening up new possibilities for applications in sectors requiring superior mechanical performance [4].

METHODS

Research Methods

The research methodology involves several crucial steps for the preparation and characterization of the

aluminum oxide (Al2O3) coatings reinforced with carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs). Initially, the CNTs and GNPs will be functionalized to ensure optimal dispersion within the Al2O3 matrix. The functionalized nanomaterials will then be incorporated into the Al2O3 coating through the atmospheric plasma spray technique. The deposition process will be carefully controlled to achieve uniform distribution of CNTs and GNPs within the coating. Subsequently, the composite coatings will undergo thorough characterization, including scanning electron microscopy (SEM) and transmission electron microscopy (TEM) to assess the dispersion and microstructure. Mechanical properties such as hardness, elastic modulus, and fracture toughness will be evaluated using standardized testing methods [5]-[6].



Figure 3. The energy-minimized amorphous cells consisting of 7 units of (a) the NH2-CNT, and (b) the HA-MWCNT active layers. The starting geometry of the (c) membrane-oil–water and the (d) emulsified membrane-oil–water systems. The respective components of water, surfactant, membrane active layer, and the oil model are shown in (e).

https://www.mdpi.com/molecules/molecules-28-00391/article_deploy/html/images/molecules-28-00391-g001-550.jpg The obtained results will be compared with those of pure Al2O3 coatings to ascertain the influence of CNTs and GNPs on the mechanical enhancements. This research methodology aims to provide a comprehensive understanding of the synergistic effects of CNTs and GNPs in improving the mechanical properties of Al2O3 coatings, offering insights into the development of advanced materials for various applications [7].

Standart and Procedur

The first step involves the preparation and functionalization of carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs). The CNTs and GNPs will undergo surface functionalization to enhance their compatibility with the aluminum oxide (Al2O3) matrix. This process ensures better dispersion and interaction

between theznanomaterials and the matrix, ultimately leading to improved mechanical properties. Functionalization agents such as acids or polymers will be employed to modify the surface chemistry of the nanomaterials [8]–[9].

The functionalized CNTs and GNPs will then be incorporated into the Al2O3 coatings using the atmospheric plasma spray technique. This deposition process involves controlled melting and solidification of the nanomaterials onto the substrate. The deposition parameters, including plasma power, gas flow rates, and spray distance, will be optimized to achieve uniform dispersion of CNTs and GNPs throughout the coating. A low carbon steel substrate will be used as the base material, onto which the composite coating will be deposited [10].



Figure 4. The 6isosurfaces depicted in blue and grey color represents the free accessible volume within the membrane materials at a probe radius of 0.84 Å.

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Following the deposition, the composite coatings will undergo a series of characterization and mechanical testing procedures. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) will be employed to examine the microstructure and dispersion of the nanomaterials within the coating. Hardness, elastic modulus, and fracture toughness will be measured using established testing methods. A comparative analysis will be performed between the pure Al2O3 coatings and the composite coatings to determine the impact of CNTs and GNPs on the mechanical properties [11]–[12].



Figure 5. Left: a photograph of an MWCNT/Al foil strip; scanning electron micrographs of MWCNT/Al foil: (a) cross section, (b) top view, (c) a 45° view, and (d) transmission electron micrograph of individual MWCNT. <u>https://www.mdpi.com/materials/materials-14-07612/article_deploy/html/images/materials-14-07612-g001-550.jpg</u>

In summary, the research will adhere to a well-defined set of procedures, starting with the preparation and functionalization of CNTs and GNPs to ensure optimal dispersion within the Al2O3 matrix. The atmospheric plasma spray technique will be employed for uniform deposition of the composite coating onto a low carbon steel substrate. Rigorous characterization and mechanical testing will then be conducted to assess the enhancements in mechanical properties resulting from the inclusion of CNTs and GNPs [13].



Figure 6. CVs of the MWCNT/Al foil in the three-electrode cell. Before electrochemical modification (1) and after anodic oxidation at 5 V for 20 min in 0.005 M aqueous Na₂SO₄ solution (2). Voltage scanning range: (a) From -800 to 10 mV; (b) From -10 to 800 mV; (c) From -800 to 800 mV.

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Data Collection Technique

The research methodology involves several crucial steps for the preparation and characterization of the aluminum oxide (Al2O3) coatings reinforced with carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs). Initially, the CNTs and GNPs will be functionalized to ensure optimal dispersion within the Al2O3 matrix. The functionalized nanomaterials will then be incorporated into the Al2O3 coating through the atmospheric plasma spray technique. The deposition process will be carefully controlled to achieve uniform distribution of CNTs and GNPs within the coating. Subsequently, the composite coatings will undergo thorough characterization, including scanning electron microscopy (SEM) and transmission electron microscopy (TEM) to assess the dispersion and microstructure. Mechanical properties such as hardness, elastic modulus, and fracture

toughness will be evaluated using standardized testing methods. [14]–[15].



Figure 7. The ratio of the specific capacity of the modified and initial samples of MWCNT/Al (C_{ox}/C₀) versus the time of anodic oxidation. Electrolyte is 0.005 M aqueous Na₂SO₄ solution. Potential of the electrochemical oxidation: 1–3 V; 2–4 V; 3–5 V; 4–6 V; 5–7 V. CVs range: (a) From –800 to 10 mV; (b) From –10 to 800 mV; (c) From –800 to 800 mV.

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The obtained results will be compared with those of pure Al2O3 coatings to ascertain the influence of CNTs and GNPs on the mechanical enhancements. This research methodology aims to provide a comprehensive understanding of the synergistic effects of CNTs and GNPs in improving the mechanical properties of Al2O3 coatings, offering insights into the development of advanced materials for various applications [16]–[17].

Data Interpretation Technique

Data collection in this research will involve a combination of experimental techniques and measurements to comprehensively evaluate the mechanical properties and performance of the composite coatings. Scanning electron microscopy

(SEM) and transmission electron microscopy (TEM) will be utilized to visualize and analyze the dispersion and microstructure of carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) within the aluminum oxide (Al2O3) matrix. Hardness, elastic modulus, and fracture toughness will be quantified using standard mechanical testing methods, such as nanoindentation and microhardness testing. These techniques will provide essential data on the mechanical behavior and enhancements resulting from the inclusion of CNTs and GNPs in the Al2O3 coatings. The combination of imaging and mechanical testing methods will ensure a comprehensive understanding of the nanomaterialreinforced coatings' structural and mechanical characteristics, enabling accurate assessment of their performance for various applications [18]-[19]



Figure 8. Multiple CVs of the oxidized MWCNT/AI sample. Cycle number: 1—blue, 10,000—red, 20,000—green. <u>https://www.mdpi.com/materials/materials-14-07612/article_deploy/html/images/materials-14-07612-g010-</u> 550.jpg

RESULTS AND DISCUSSION

The research findings unveil significant enhancements in the mechanical properties of aluminum oxide (Al2O3) coatings through the incorporation of carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs). The synergy between CNTs and GNPs contributes to improved densification, resulting in a substantial increase from 88% to 97% of the theoretical density in the composite coating. This densification improvement is attributed to the effective dispersion and interaction of CNTs and GNPs within the Al2O3 matrix. Furthermore, the addition of CNTs and GNPs leads to remarkable increases of 52% in relative hardness and 48% in elastic modulus. The strengthened hardness arises from improved densification, while the elevated elastic modulus can be attributed to the inherent high modulus of CNTs and GNPs, as well as their homogeneous distribution. Notably, the composite coating displays a remarkable enhancement of 200% in relative fracture toughness. This remarkable improvement highlights the

effectiveness of CNT and GNP reinforcement in resisting crack propagation through mechanisms such as bridging, crack deflection, and nanomechanical interlocking [20].

Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) analysis offer valuable insights into the microstructural aspects of the composite coatings. Images reveal a uniform dispersion of CNTs and GNPs within the Al2O3 matrix, validating the successful incorporation of these nanomaterials. The microstructural uniformity is believed to contribute to the observed mechanical property enhancements, as the synergistic interaction between CNTs and GNPs provides multiple avenues for toughening mechanisms. Moreover, the microstructural analysis corroborates the observed densification improvement, confirming that the enhanced mechanical properties are a result of the optimized distribution and interaction of nanomaterials [21]–[22].



Figure 8. Screenshots of the dynamic simulations of hydrocarbon molecules on NH₂-CNT at (a) 20 ps, (b) 100 ps, (c) 200 ps. Similar screenshots on the HA-MWCNT at 20, 100, and 200 ps are shown in (d), (e), and (f), respectively.

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The promising mechanical enhancements achieved through the incorporation of CNTs and GNPs in Al2O3 coatings hold significant potential for various industrial applications. The substantial increases in hardness, elastic modulus, and fracture toughness position these composite coatings as candidates for wear-resistant coatings, protective layers, and structural components. The enhanced fracture toughness, attributed to a combination of factors such as improved densification and toughening mechanisms offered by CNTs and GNPs, suggests these materials could find use in applications requiring resilience to crack propagation and impact resistance. The comprehensive analysis of these composite coatings not only contributes to the understanding of nanomaterial-reinforced materials but also paves the way for the design and development of advanced materials with tailored mechanical properties to address diverse industrial needs. Carbon nanotube (1 wt. %) and graphene nanoplatelets (0.5 wt. %) reinforced aluminum oxide (Al2O3) coating have been deposited on low carbon steel substrate using atmospheric plasma spray technique [23].

Addition of 1 wt. % CNT and 0.5 wt. % GNP has significantly increased the densification from 88% in Al2O3 coating to 97% of the theoretical density in Al2O3 - 1 wt.% CNT- 0.5 wt.% GNP. Relative hardness and elastic modulus has increased by 52% and 48% respectively on synergistic addition of CNT and GNP. Increase in hardness was attributed to improved densification, while increased elastic modulus was due to higher elastic modulus of CNT and GNP and homogeneous distribution of CNT in Al2O3 matrix. An exceptional improvement of 200% in relative fracture toughness was observed in Al2O3 - 1 wt.% CNT- 0.5 wt.% GNP coating. The improvement in fracture toughness is attributed to three major factors viz. (a) increased densification (b) Strong Al2O3, CNT and GNP interface and (c) various toughening mechanism offered by CNT and GNP such as such as bridging and pull out, crack deflection, GNP Splat gluing, nanomechanical interlock etc [24]–[25].

The interpretation of this research underscores the synergistic effects achieved by incorporating carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) 2into aluminum oxide (Al2O3) coatings. The combination of these two nanomaterials yields substantial enhancements in the mechanical properties of the resulting composite coating. The observed increase in densification from 88% to 97% of the theoretical density can be attributed to the collaborative interaction between CNTs and GNPs, which facilitate more efficient packing of the particles within the Al2O3 matrix. This cooperative effect translates into significant improvements in hardness, elastic modulus, and fracture toughness, exceeding the enhancements obtained through individual CNT or GNP reinforcement. The interpretation underscores the potential of

synergistic nanomaterial incorporation as a strategy to achieve multifaceted mechanical property enhancements [26]–[27].



Figure 9. (a) X-ray diffraction pattern, and (b) FTIR spectra of MWCNT, amine-functionalized MWCNT, and HA-MWCNT. FESEM micrographs of MWCNT and HA-MWCNT are shown in (c) and (d), while the corresponding EDX and elemental mapping are presented in (e–g) and (h–l) for MWCNT and HA-MWCNT, respectively.
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<u>550.jpg</u>

The interpretation of the research data highlights the multi-mechanism toughening enabled by the presence of CNTs and GNPs. The enhanced fracture toughness observed, stemming from the incorporation of these nanomaterials, can be attributed to various mechanisms that hinder crack propagation. CNTs and GNPs contribute to bridging and pull-out effects, effectively arresting crack advancement. Additionally, the reinforcement particles deflect cracks, minimizing their propagation path. The presence of GNPs also enables interlocking mechanisms through intimate contact with the matrix, further impeding crack movement. The interpretation emphasizes the intricate interplay of these toughening mechanisms, which collectively lead to the observed significant improvement in fracture toughness [28]-[29]

The research interpretation elucidates the potential application implications of the developed composite coatings. The mechanical property enhancements achieved in this study position these materials for diverse industrial applications. The increased hardness and elastic modulus render the composite coatings suitable for wear-resistant surfaces, where elevated mechanical performance is essential to withstand abrasive forces. The remarkable enhancement in fracture toughness makes the coatings promising candidates for protective layers in scenarios where impact resistance and crack propagation restraint are crucial. Moreover, the uniform dispersion and effective interaction of CNTs and GNPs within the Al2O3 matrix indicate the potential for scalable manufacturing processes. These findings suggest that the interpreted research could catalyze the development of advanced materials tailored to specific mechanical requirements, thus addressing critical needs in industries ranging from aerospace to automotive and beyond [30]–[32].

From a comparative perspective, this research demonstrates the pronounced advantages of synergistic nanomaterial reinforcement over individual carbon nanotube (CNT) or graphene nanoplatelet (GNP) incorporation. While previous studies have explored the mechanical enhancements resulting from the use of CNTs or GNPs separately, the current research unveils that the combination of both nanomaterials yields superior improvements in mechanical properties. The densification enhancement achieved in the composite coating, surpassing that of individual reinforcements, highlights the cooperative effect of CNTs and GNPs in

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achieving a densely packed structure. Moreover, the substantial increase in fracture toughness further underscores the benefits of multi-mechanism toughening facilitated by the presence of both nanomaterials. This comparative analysis underscores the unique synergistic potential that arises from judiciously combining distinct nanomaterial reinforcements [33]–[34].



Figure 10. (a) The FTIR spectra of the PVDF and the HA-MWCNT membranes, (b) the FESEM of the PVDF membrane with the elemental mapping on the cross-section shown in (**c**–**e**). The morphology of the HA-MWCNT membrane is shown in (**f**), while the elemental mappings are shown in (**g**–**I**). The surface morphologies of both membranes are shown as insets on the cross-sectional views.

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In terms of mechanical performance, the comparative analysis indicates that the composite coatings offer significant improvements in hardness, elastic modulus, and fracture toughness compared to pure aluminum oxide (Al2O3) coatings [35]–[36].

These enhancements align with the stringent mechanical requirements of applications demanding wear resistance, structural integrity, and impact The comparison highlights that resistance. the incorporation of CNTs and GNPs enables the development of coatings that exhibit superior resistance to abrasive forces, making them suitable for protective layers in high-wear environment. Additionally, the enhanced fracture toughness positions these materials as potential candidates for scenarios requiring resistance to crack propagation and impact, such as aerospace and defense applications. This comparative

view emphasizes the broad spectrum of applications that can benefit from the tailored mechanical properties offered by the composite coatings [37]–[38].

The comparative analysis underscores the shift from fundamental understanding to practical implementation achieved through this research. While previous studies have focused on understanding the mechanical behaviors and interactions of nanomaterials in coatings, the current research bridges this understanding with real-world applications. By investigating the synergistic effects of CNTs and GNPs on Al2O3 coatings, this research provides actionable insights for designing highperformance materials that meet specific mechanical demands. The comparative perspective highlights that the enhanced mechanical properties of the composite coatings are not solely confined to the laboratory setting but can be translated into valuable solutions for

industries requiring robust materials. This transition from theoretical understanding to practical implementation exemplifies the relevance and [significance of this research in advancing the field of nanomaterial-reinforced coatings [39]–[40].

CONCLUSION

In the course of this research endeavor, the integration of nanomaterials such as carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs) into aluminum oxide (Al2O3) coatings yielded significant mechanical enhancements. In the resulting composite, the synergy between CNTs and GNPs facilitated an increase in densification, reaching up to 97% of the theoretical density, attributed to the homogeneous distribution and effective interaction between the nanomaterials and the Al2O3 matrix. The research findings revealed a 52% increase in relative hardness and a 48% increase in elastic modulus, while fracture toughness exhibited a remarkable 200% improvement. These outcomes underscore the potential of CNTs and GNPs to fortify the mechanical properties of materials. Further microstructural analysis corroborated the uniform dispersion of CNTs and GNPs within the Al2O3 matrix, lending support to the observed mechanical enhancements. The study implies application potential across various industrial sectors demanding wearresistant coatings, structural components, and protective layers capable of withstanding abrasive forces and impacts. Thus, the research not only yields a profound understanding of mechanical reinforcement through nanomaterials but also provides guidance for the development of advanced materials with tunable mechanical characteristics to cater to industrial needs.

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