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Properties of Alumina-Titania Hybrid Nanocomposite for Metallic Coating: A Brief Review

Mahani Yusoff[1,2,*], Teo Pao Ter[2], Nor Fadhilah Ibrahim[2], Mohd Hasmizam Razali[3]

[1]Functional and Sustainable Nanomaterials Research Group, Faculty of Bioengineering and Technology, Universiti Malaysia Kelantan, 17600 Jeli, Malaysia
[2]Faculty of Bioengineering and Technology, Universiti Malaysia Kelantan, 17600 Jeli, Kelantan, Malaysia
[3]Faculty of Science and Marine Environment, Universiti Malaysia Terengganu, 21030 Kuala Nerus, Terengganu, Malaysia

***E-mail**: mahani@umk.edu.my

ABSTRACT: The hybrid nanocomposite of alumina-titania (Al_2O_3 - TiO_2) is a material with unique features that make it a good possibility for coatings that inhibit corrosion and wear. This nanocomposite is composed of Al_2O_3, TiO_2, and third-phase particles. Introducing a third phase, such as carbon-based material or oxide ceramic, into an Al_2O_3-TiO_2 composite in a ternary system drastically changes the composite's properties. These three combination materials result in nanocomposites with increased hardness, wear resistance, chemical resistance, and thermal stability. The spraying technique is commonly used to deposit Al_2O_3-TiO_2 hybrid nanocomposites to coat a metal substrate. The microstructure, porosity, and mechanical properties of the nanocomposite feedstock determine the quality of the decorated substrate. The properties of the nanocomposite can be adjusted by modifying the composition and size of the nanoparticle. Al_2O_3-TiO_2 hybrid nanocomposites with tertiary materials show great potential as a novel material with various possible uses in metallic coating. A new, unique ternary system, as well as improved coating properties in Al_2O_3-TiO_2 hybrid nanocomposites, require more exploration.

Keywords: Alumina-titania, metallic coating, nanocomposites, Bi-modal Microstructure

1. Introduction

Ceramic-composite coatings are preferred because they are heat-resistant, anticorrosive, and wear-resistant under severe pressure and temperature conditions. They are, thus, utilized to protect metallic materials under harsh circumstances, such as combustion engines, storage tanks, hydropower plants, and turbine components. Two important considerations for ceramics-composite coating are the intrinsic properties of ceramics and the method of applying the layer on the substrate. Different coating processes have been developed to use the ceramic coating onto the substrate, such as plasma spray, vacuum deposition, and chemical vapor deposition. Typically, ceramic composite coating particles are sprayed onto the substrate to create two-dimensional layers.

Composite matrix composites (CMCs) consist of ceramic as a matrix phase in the composites, while the reinforcement can be any material, whether it is metals, ceramic, or polymer. Recently, the development of ceramics composite coating focuses on producing ceramics/ceramics and ceramics/metal to replace monolithic ceramic. CMCs have a greater elastic modulus and can withstand higher plastic deformation than monolithic ceramics. However, the structural heterogeneity of CMCs commonly produces defects such as voids and cracks and leads to reduced corrosion resistance. Therefore, the microstructure, size, shape, and distribution of the phases are altered into ultra-fine or nanostructure materials. Ceramic-based nanocomposites (CMCs) are applied to improve these favorable properties. The CMNCs exhibit firm adhesion to metallic substrates, good tribological properties, and high wear and corrosion resistance. In addition, CMNCs coating presents not only antioxidative properties but unique features such as flame retardancy, excellent thermal stability, phase stability, and light-weight and smooth-surface coating. The weak contact between the layers of steel and ceramic causes coating ceramic on metal a challenging task. The complex mechanism of adhesive bonding between ceramic-metal substrates is addressed through understanding the substrate surface topography and chemical behavior of ceramic-metal. Under appropriate chemical and thermodynamic conditions, metal-to-metal interaction between the atoms in the base metal and metallic ions in the ceramic is the reason for the excellent binding of ceramics to metals. In creating high-performance oxide layers, the oxide phase is the main component for ceramic coating. Most oxide phases, such as chromium oxide, titanium oxide, aluminum oxide, and zirconium oxide, have a melting point of up to 2000°C, indicating they are stable at high temperatures. They also can be directly thermally sprayed without a protective atmosphere. Several oxide ceramics and oxide-CMCs for metallic coatings are shown in **Table 1**.

Table 1: Various oxide ceramics and oxide-CMC coatings on the metallic substrates

Oxide and oxide-CMCs system	Coating techniques	Coating Hardness (HV)	Coefficient of Friction	Substrate	Reference
Al_2O_3-TiO_2	Plasma spray	816	-	Alloy steel	[1]
	Plasma spray and laser remelting	630	-	Carbon steel	[2]
	Dipping	-	0.4 - 06	Stainless steel	[3]
Al_2O_3- Zr_2O_3	Plasma spray	-	-	Carbon steel	[4]
Al_2O_3-NiAl	HVOF	-	0.75	Stainless steel	[5]
Al_2O_3-YSZ	Plasma spray	-	0.21	Alloy steel	[6]
TiO_2-SiC	Thermal spray	958	-	Ti alloy	[7]
TiO_2-Zr_2O_3	Plasma spray	317	-	Al alloy	[8]
	Sol-gel spin	300	-	Low carbon steel	[9]
WO_3-Al_2O_3	Ultrasound-enhanced Micro-Arc Oxidation	1025	-	Al alloy	[10]
Cr_2O_3	HVOF	1300	-	Stainless steel	[11]

Alumina-titania (Al_2O_3-TiO_2) composites are a unique material with a high melting point and chemical inertness, permitting them to retain mechanical qualities like hardness and strength even at temperatures as high as 1100 °C. Therefore, it is the appropriate material for coatings used in corrosive and high-temperature situations. However, when deposited using thermal spray techniques, the coating structure may develop voids and cracks because of agglomerated nanostructure and delamination. As a result, Al_2O_3-TiO_2 coatings are undesirable for applications where specific types of degradation are of concern and susceptible to certain types of degradation environments. Attempts are made to produce these materials as nanocomposites with a hybridization strategy. The advancements in Al_2O_3-TiO_2 composite coatings, unique characteristics of Al_2O_3-TiO_2 hybrid nanocomposite coatings, and their applicability on a range of metallic substrates are all described in this review paper.

2. Al_2O_3-TiO_2 Composite

Al_2O_3-TiO_2 is a two-phase composite in which TiO_2 reinforces the Al_2O_3 matrix. Al_2O_3-TiO_2 composites have gained recognition for their exceptional toughness, low thermal expansion, low thermal conductivity, and low porosity. Electrical insulation and wear-resistant coating are made possible by the high hardness of Al_2O_3. Al_2O_3, which contains about 3 wt% TiO_2, is often used as a coating for wear resistance applications. One of the distinguishing morphology features of Al_2O_3-TiO_2 coating materials is the interlocking microstructure provided by TiO_2 in Al_2O_3-TiO_2 composite. TiO_2 originating has a low melting point, and they could be dispersed throughout the microstructure along with Al_2O_3 grains. Inhibition of the Al_2O_3 agglomeration by TiO_2 is achieved through partially embedded upon the Al_2O_3 pores. This influences the overall mechanical and thermal properties of the composite. In an Al_2O_3-TiO_2 system, two matrix phases are typically observed: γ-Al_2O_3 and α-Al_2O_3. The higher nucleation energy of γ-Al_2O_3 than α-Al_2O_3 accounts for its more dominant position than α-Al_2O_3. It has been reported that at 1000°C to 1100°C, the transition γ-Al_2O_3 to α-Al_2O_3 [12] occurred with a drastic reduction in the surface area [13], increment of hardness [14] and wear resistance [15].

Achieving the desired coating characteristics requires modifying the phase composition, porosity, and particle size of Al_2O_3-TiO_2. Several studies have been carried out on these considerations. The total Al_2O_3 and TiO_2 contents have been indicated to influence the bi-modal microstructure by producing a dense oxide layer [16]. However, the addition of high TiO_2 content was found to reduce the composite hardness while maintaining the toughness [17, 18] [19], [20]. Luo et al., 2017 [21] and Richter et al., 2019 [22] investigated the impact of 40 wt% TiO_2 addition. They found that in this composition, Al_2TiO_5 (aluminium titanate) dramatically influences the densification, hardness, and elastic modulus of the composite coating. As is frequently observed for Al_2O_3-TiO_2 composites with higher than 30 wt% TiO_2, controlling the Al_2TiO_5 phase in composite coatings must be addressed [23]. It is well known that a coating with lesser porosity is influenced by starting materials with small particle sizes. On the metallic substrate, lower porosity typically results in good distribution and adhesion to surfaces. According to research on incorporating nanostructured Al_2O_3-TiO_2 feedstock for plasma spray, the obtained mechanical properties are similar to the submicron-sized particles, which was very much related to its microstructure [24]. In addition, significantly improved wear resistance was observed for Al_2O_3-TiO_2 nanocomposite compared to Al_2O3-TiO_2 microcomposite [25]. In addition, considerable progress on wear resistance is observed for Al_2O_3-TiO_2 nanocomposite compared to Al_2O_3-TiO_2 micro-composite [25]. The amount of build-up porosity of the coating comes from the integration of mechanical interlocking and diffusion bonding [26]. However, nanostructured Al_2O_3-TiO_2 commonly featured an agglomeration, which contributes to limiting the flowability upon spraying.

3. Al_2O_3-TiO_2 Nanostructure Hybridization

Ceramic composite coating is composed of combined materials with different properties. Numerous works have been done to integrate the ceramics mixed layer into nanocomposite with superior properties. Coatings made of nanostructured Al_2O_3-TiO_2 have a uniform microstructure and excellent interfacial adhesion between metal substrates [27-31]. Their morphology is composed of the TiO_2 particles that are partially melted and embedded in Al_2O_3 nanocrystalline particles, resulting in a bi-modal microstructure as shown in Figure 1. This microstructure has a different characteristic by following the ability to bond chemically and the materials' dispersion behavior. Most developed ceramic hybrid nanocomposite coatings have reported improved properties by adding a third oxide phase in an Aln an Al_2O_3-TiO_2 composite.

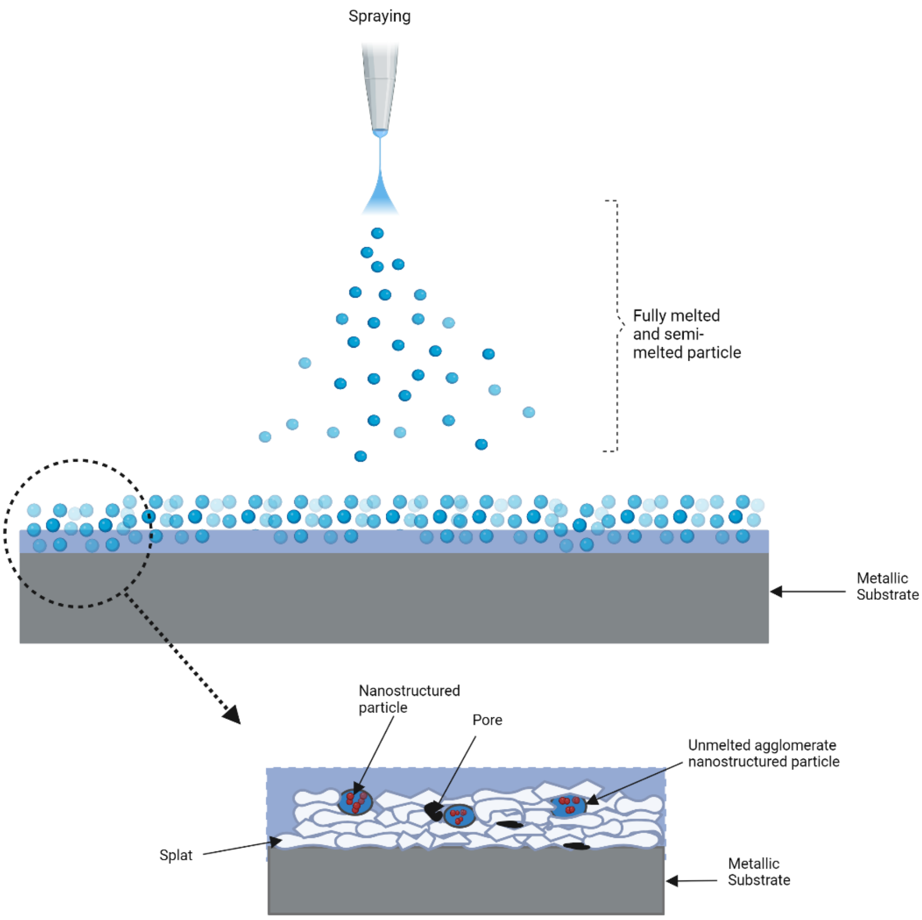

Figure 1: Typical schematic bi-modal coating microstructure of sprayed coatings made of nanostructured ceramic composite

3.1 Al_2O_3-TiO_2-CNT

Carbon nanotubes (CNTs) are nanomaterials made of cylindrical molecules formed by rolling a carbon atom in a hexagonal pattern and appear as single CNT and multi-walled CNT (MWCNT). CNT has been widely utilized in small portions in the composite as a photocatalyst for wastewater treatment [32-35] and nanofluids in heat transfer applications [36, 37]. CNT is not only utilized as reinforcement for strengthening purposes but also acts as a lubricant and reduces the coefficient of friction [38]. Simultaneous effect on CNT making them favorably long been used reinforcement in Al_2O_3 [39-42], ZrO_2 [43-46], zirconia toughened Al_2O_3 [47, 48] ceramic coatings.

Several studies have been done on the performance of coated layers containing Al_2O_3-TiO_2 with CNT. Basha et al., [49] prepared different concentrations of CNT (2, 4, and 6 wt%) to study the influence of CNT on the performance of microhardness, porosity as well as the surface roughness of the composite coating deposited by air-plasma spray (APS). The wear performance of the layer is dependent on the percentage of the porosity-coated substrate. Adding 1 wt % MWCNT on Al_2O_3-TiO_2 feedstock results in good wear properties because it has fewer cracks observed [50]. In corrosion protection, using 1.5 wt% and 3.0 wt% CNT loadings contributes to a denser coating layer, which prevents further oxidation under a harsh environment [51]. Further, synergistic effects on electromagnetic wave absorption and interference with good flexural strength were obtained as low as 5 wt% CNT content, enabling them to be alternatively used in microwave applications [52].

3.2 Al_2O_3-TiO_2-graphene

Graphene is a two-dimensional nano-layered structure of carbon atoms with ultra-thin thickness and excellent mechanical properties superior to graphite and CNT. Graphene appears in various forms: monolayer graphene, graphene nanoplatelets (GNPs), graphene oxide (GO), and reduced graphene oxide (rGO). They are extensively used in all various matrix material types for composite coatings due to their exceptional strength. A small amount of graphene added to a ceramic composite enhances the interfacial area and improves the composite's overall properties [53-56]. By strengthening the interfacial region, nanoparticles significantly enhance the mechanical, thermal, and electrical properties of the composite.

Most research on Al_2O_3-TiO_2-filled graphene nanocomposite has concentrated on graphene, GNPs, and rGO as fillers. The toughening mechanism of these fillers had a considerable impact on the mechanical properties and wear resistance of the ceramic composite coating. Graphene with different contents (0, 3.0, 6.0, 9.0, and 12.0 wt%) was investigated by Wang et al. [57]. They proposed that wear efficiency decreases when graphene content exceeds 6 wt% graphene, despite hardness does not change appreciably. Verma et al. [58] fabricated plasma-sprayed Al_2O_3-TiO_2-GNPs with 0.5, 1.0, 1.5, and 2.0 wt% GNPs, revealing that the mechanical and tribological properties highly depend on GNPs composition. The nanocomposite's outstanding hardness and wear performance on seawater is demonstrated by 1.5 wt% GNPs, which originated from good dispersion, less agglomeration, smoother surface, and lubricating effect of GNPs. Moreover, the GNPs get separated from the substrate, stuck to the worn surface, and form a lubricating coating that could contribute to the reduction of friction coefficient [59]. Recently, the development of Al_2O_3-TiO_2-rGO nanocomposite utilizing different amounts ranging 0.5 – 1.5 wt% of rGO was demonstrated by Srikanth and Bolleddu [60]. According to the research findings, elevating the concentration of rGO within the composite coatings leads to a gradual decrease in porosity. Moreover, adding rGO to coatings strengthens them against fractures by creating a strong bond between the coating layer, effectively retarding the crack propagation.

Most studies on Al_2O_3-TiO_2 hybrid nanocomposite do not include the mechanism for improving mechanical properties such as fracture toughness and interfacial bonding between graphene, Al_2O_3, and TiO_2. Grain refinement strengthening, Orowan strengthening, dislocation strengthening by CTE mismatch, and load transfer matrix reinforcement essentially attribute the cracking and pull-out of graphene to its strengthening effect in the Al_2O_3 matrix [61]. Moreover, strong π–π interactions and considerable Van der Waals force cause graphene to build up and allow it to tend to reorganize [62]. Therefore, an emphasis on the possible toughening mechanisms that may operate in this hybrid nanocomposite and the deposition mechanism on metallic substrate should be explored.

3.3 Al_2O_3-TiO_2-Cr_2O_3

Chromia (Cr_2O_3) is an oxide ceramic with excellent chemicals and wear resistance, which makes it suitable for corrosion protection. These properties are related to the chemically inert α-Cr_2O_3 that leads to more stable composite coating [63]. Cr_2O_3 particles incorporated composite to coat metallic coatings had a considerable hardening effect, allowing them to minimize more significant stresses and resist wear adhesion. Grimm et al. [64] fabricated a thermally sprayed coating feedstock of an Al_2O_3-TiO_2-Cr_2O_3 composite [65]. They reported the composite consists of α-Al_2O_3 and γ-Al_2O_3, rutile TiO_2, and Cr_2O_3 after several modifications on spraying parameters. The metastable phase of Cr_2O_3 does not occur as they are being reverted during cooling from oxidation of Cr_2O_3 to chromium trioxide (CrO_3) [66-68]. These phases are responsible for internal structure changes and phase transformation during heating and cooling. In a further investigation, they demonstrated that in-situ atmospheric plasma spraying with high content Cr_2O_3 (35 wt%) reduced the wear rate of Al_2O_3-TiO_2-Cr_2O_3 to half that of pure Al_2O_3 [69].

3.4 Al_2O_3-TiO_2-Y_2O_3

Yttria (Y_2O_3) is an oxide ceramic commonly used as reinforcement in composites due to its high strength, hardness, melting point, and thermal conductivity. Early work on ternary Al_2O_3-TiO_2-Y_2O_3 systems focused on structural and phase transition synthesized by physical methods [70-72]. Mehar et al. [73] developed plasma sprayed Al_2O_3-TiO_2-Y_2O_3 composite focusing on its tribological properties. They compared the Al_2O_3-TiO_2 and Al_2O_3-TiO_2-Y_2O_3 composites. The composite containing Y_2O_3 resulted in a significant reduction of the coefficient of friction and wear rate because of the combined effects of enhanced fracture toughness, stabilized α-Al_2O_3 phase, and Ti magneli phase oxides. It is important to take into account depositing coating using thermal spraying because the isothermal area occurred between 1550 and 1400°C [74]. In addition, the co-existence of the α- and γ-Al_2O_3 phases is noticed, suggesting that the pressure from APS is insufficient to convert Al_2O_3 into a fully stable state.

3.5 Al_2O_3-TiO_2-Zr_2O

Zirconia (Zr_2O) is an oxide ceramic with good fracture toughness, high strength, and excellent wear resistance. Zr_2O particles are utilized as a reinforced phase in various ceramic matrices potentially used for thermal barrier coating due to good fracture toughness [70, 75], hardness [76], low porosity [75, 77], and thermally stable [78]. Gao et al. prepared Al_2O_3-TiO_2-Zr_2O composite coating using plasma spray. They observed that the Al_2O_3–ZrO_2–TiO_2 coating resulted in a more compact layer with fewer voids and microcracks. The composite shows a better response on adhesion and wear testing than the Al_2O_3–ZrO_2 composite. Sathish et al. [79] demonstrated the wear and corrosion performance using this composite for titanium alloy coating in biomedical applications. Three ZrO_2 compositions have been studied: 2, 5, 8%. The result showed that a lower ZrO_2 (2%) composition led to the lowest wear rate, strongly related to phase formation and the melting rate of consolidated coating particles.

4. Challenges and future works

The Al_2O_3-TiO_2 hybrid nanocomposite coating faces a few obstacles, including uniform nanoparticle dispersion, shape and size control, improved interfacial adhesion, and the development of economical synthesis methods. The research on these factors is limited because of the lack of detailed information on phase transition and adequate isothermal regions. This information is useful for depositing feedstock material using the plasma spray technique.

Optimizing the properties of the nanocomposite requires achieving a homogenous distribution of particles. This can be difficult due to the nanocomposite particles' tendency to agglomerate. Agglomeration led to greater porosity, which exposed the substrate to the environment by producing defects such as voids and cracks. Furthermore, the characteristics of the nanocomposite can be strongly influenced by the size and shape of the particles. It is crucial for controlling these components to tailor the nanocomposite for specific use. The interfacial adhesion of the particles significantly influences the performance of the nanocomposite. Weak interfacial adhesion can cause delamination and lower mechanical properties. The current Al_2O_3-TiO_2 hybrid nanocomposite synthesis techniques, such as the sol-gel process, can be laborious. Ensuring the large-scale production of these nanocomposites requires creating more economical and efficient synthesis techniques. Further development and broader application of Al_2O_3-TiO_2 hybrid nanocomposite in coating applications would be made possible by overcoming these barriers.

Developing a hybrid nanocomposite coating, which combines Al_2O_3 and TiO_2 with other materials can lead to notable improvements in component performance. This, in turn, can result in a longer lifespan for the components and a reduction in maintenance costs. Moreover, hybrid nanocomposites often allow for the optimized use of materials that may exploit the unique properties of each material while minimizing resource consumption. It is crucial to balance the economic benefits of advanced nanocomposite coatings with responsible and sustainable resource usage to ensure their long-term success and acceptance in various industries.

5. Conclusions

This review highlights the Al_2O_3-TiO_2 hybrid nanocomposite as a promising material for coating applications due to its unique combination properties, including high hardness, excellent chemical resistance, and good thermal stability. Developing Al_2O_3-TiO_2 hybrid nanocomposite was observed based on the selected third phase, which is alternatively used to coat ferrous and non-ferrous alloy substrates. Incorporating CNT, graphene, Cr_2O_3, Y_2O_3, and Zr_2O into the Al_2O_3-TiO_2 composite makes up a ternary system that, in small quantity, further enhances its mechanical properties, chemical resistance, and thermal conductivity. These improved properties are related to the phase transformation, microstructure, and dispersion of particles inside the nanocomposite. Challenges include uniform nanoparticle dispersion, shape, and size control, improved interfacial adhesion, and the development of economical synthesis methods.

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Conflict of Interest

The authors declare that they have no conflict of interest.

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