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Adhesion of Magnetron Sputtering coatings on Acrylonitrile Butadiene Styrene: A systematic review

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Abstract. *This work presents a systematic review conducted to assess the adhesion properties of coatings deposited by Magnetron Sputtering on Acrylonitrile Butadiene Styrene (ABS). Employing the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) methodology, relevant literature was searched across prominent platforms, including Scopus, Springer, Taylor and Francis, Wiley, and Web of Science. A comprehensive search yielded 122 articles based on specific keywords, out of which 23 were excluded due to being review articles, and 7 were eliminated as duplicates. A total of 92 articles were then evaluated against predetermined eligibility criteria, leading to the selection of 12 articles for inclusion in the final review. The synthesis of these articles focused on examining the impact of deposition parameters' variations and the utilization of interlayers on coating adhesion. Through a rigorous systematic approach, this review provides a comprehensive overview of the current knowledge and trends in the field, offering valuable insights for future research and industrial applications.*

Keywords: *Adhesion, Coating, Magnetron Sputtering, Acrylonitrile Butadiene Styrene, Systematic Review*

1. INTRODUCTION

The adhesion of coatings deposited on polymeric substrates is a topic of great interest in the field of materials science and engineering. The use of coatings is an effective way to improve the mechanical, chemical, and thermal properties of polymeric materials, making them suitable for a wide range of applications in several industries (Lan *et al.*, 2020).

Physical vapor deposition (PVD) is notable in this scenario for its ability to deposit both organic and inorganic materials, making it highly versatile. Additionally, PVD is environmentally friendly, as it minimizes waste and avoids the use of harmful chemicals. Furthermore, its capacity to deposit materials at low temperatures is particularly advantageous for polymers, which are susceptible to melting or degrading at high temperatures (Makhlouf, 2011).

The PVD methods encompass various vacuum deposition techniques utilized to produce thin coatings. Among them, Magnetron Sputtering stands out for its ability to achieve high deposition rates, film purity, and coating adhesion (Hassan, 2018; Tudose *et al.*, 2019). While the process parameters for coating metals like steel are already established (Blesman *et al.*, 2017; Vega-Morón *et al.*, 2018; Recco *et al.*, 2007; Lattemann *et al.*, 2006; Deng *et al.*, 2020; Chen and Duh, 1992), further systematic research is required to optimize the technology for coating polymeric materials. This includes developing surface preparation techniques and deposition parameters to enhance adhesion.

In this scenario, the main objective of this study is to systematically review the literature on the adhesion of coatings deposited by magnetron sputtering on acrylonitrile butadieno styrene (ABS) and to evaluate the quality of the existing research using the Preferred Reporting Items for Systematic Reviews and MetaAnalyses (PRISMA) methodology. The study will provide an overview of the current state of knowledge on adhesion from the results obtained by systematic searches. The analysis will highlight the factors that affect adhesion, including surface pretreatment, deposition parameters, and the use of the interlayers. The review will also identify the gaps in the literature and provide recommendations for future research.

To quantify the outcomes, the coating most studied were CrN (Sukwisute *et al.*, 2017; Zhang *et al.*, 2020; Pedrosa *et al.*, 2018), Cu (Lambaré *et al.*, 2015; Dai *et al.*, 2023; Jachowicz, 2021; Afshar and Mihut, 2020), Cu/Ti (Lambaré *et al.*, 2015), TiN (Na *et al.*, 2003; Grimberg *et al.*, 1994; Jachowicz, 2021), Al (Jachowicz, 2021; Heinß and Fietzke, 2020; De Bruyn *et al.*, 2003), TiAl (Jachowicz, 2021) and Ag (Zendehnam *et al.*, 2014).

2. METHODOLOGY

This systematic review evaluated its methodological quality by adhering to the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) statements for all of its methodology procedures (Glanville *et al.*, 2021).

2.1 Research Question

The Population, Intervention, Comparison and Outcome (PICO) methodology was used to formulate a research question prior to starting this systematic review (Schardt *et al.*, 2007). Thus, the question to be answered is: ”How to improve the adhesion of coatings deposited by Magnetron Sputtering on ABS?”

2.2 Search Strategy

2.2.1 Databases Searched

The article search was carried out independently by two researchers in four electronic databases, namely Scopus, Springer, Taylor & Francis, Wiley, and Web of Science. The search filters were used to locate scientific papers in journals written in English until July 2023.

2.2.2 Keywords Selection

Using the PICO methodology, a combination of keywords was developed. So, the string used in the search was: "("MAGNETRON SPUTTERING") AND ("ACRYLONITRILE BUTADIENE STYRENE")", limited to papers written in English.

2.2.3 Search Fields

Keywords were searched in the title, abstract, and keywords fields of the articles to ensure the relevance of the retrieved papers.

2.3 Eligibility Criteria

The criteria below were established for article inclusion:

- The coating was deposited on ABS;
- The film was deposited by the Magnetron Sputtering technique;
- Adhesion test was performed;
- Published in peer-reviewed journals;
- Written in English.

The article exclusion happened when one of the cited criteria was not accomplished.

2.4 Study Selection

The study selection process followed a two-stage screening procedure:

2.4.1 Initial Screening

Two independent reviewers screened articles based on titles and abstracts. Articles that did not meet the inclusion criteria were excluded.

2.4.2 Full-Text Review

Selected articles from the initial screening underwent a full-text review by the same two reviewers. Any disagreements were resolved through discussion, and a third reviewer was consulted if needed.

2.5 Data Extraction

A structured data extraction form was developed to collect relevant information from the selected articles, including:

- Authors and publication details;
- Key findings related to strategies for improving adhesion in Magnetron Sputtering coatings on ABS (such as pre-treatments and process parameters);

2.6 Data Synthesis

The extracted data were synthesized to identify common strategies, techniques, and findings related to enhancing coating adhesion on ABS polymer substrates using Magnetron Sputtering. A narrative synthesis approach was employed to present the results.

2.7 Discussion and Conclusion

The systematic review's findings will be discussed in the context of the research question, and conclusions will be drawn regarding the effectiveness of various strategies for improving coating adhesion on ABS polymer. Implications for future research and practical applications will also be considered.

2.8 References

A comprehensive list of the references cited in the systematic review will be provided in accordance with the PRISMA guidelines.

2.9 Reporting

The systematic review will adhere to the PRISMA guidelines for transparent and comprehensive reporting of the search strategy, study selection process, data extraction, and synthesis of findings.

By following this methodology, we aim to provide a rigorous and comprehensive assessment of the existing literature on strategies to improve coating adhesion in Magnetron Sputtering on ABS polymer, ultimately contributing to the advancement of this important field of research and industrial applications.

3. RESULTS

A total of 122 articles were identified through a keyword search described in Section 2.2.2with 76 found in Scopus, 22 in Wiley, 16 in Springer, 6 in Web of Science and 2 in Taylor & Francis . Before starting the screening stage, 7 references were removed because they were duplicates, and 23 were removed because they were review papers. Thus, 92 studies proceeded to the next step. Then, the title, abstract, and keywords of 92 articles were evaluated, of which 17 were selected for a full reading. Following this step, 6 articles were excluded because they were not relevant to the scope of work, and one were added by reference analysis. Therefore, analyses of 12 articles were performed. All these steps are described in Figure 1. The main characteristics of the coating deposition parameters are summarized in Tab. 1.

Regarding the bibliometric analysis, Figure 2 shows the publications year. From the first to the last publication, there is a 30-year interval. Analyzing the results in 10-year increments, in the range of 1994-2003, 3 articles were published, related to the years 2004-2013, 1 article, and in the 2014-2023 period, 8 articles were published. This indicates an increasing trend of research in the field of coatings deposited by Magnetron Sputtering on ABS.

A comprehensive literature review reveals that there are no significant disparities in the number of publications by country. An analysis of a diverse set of articles shows that Germany, China, Singapore, and France have two publications in the field. Following them, with one publication each, are South Korea, Israel, Portugal, Iran, Thailand, the United States of America, Poland, and Belgium. These findings highlight the global distribution of research efforts in the field of surface engineering.

In the academic research field, universities stand out in terms of the number of publications; however, two works were published as a result of cooperation between industry/private research institutes and universities. Examining the concentration areas in which the authors operate, all the works were developed in engineering centers. Nevertheless, three works featured multidisciplinary cooperation between different centers, namely: physics and materials engineering, physics and chemical engineering, and physics and manufacturing engineering.

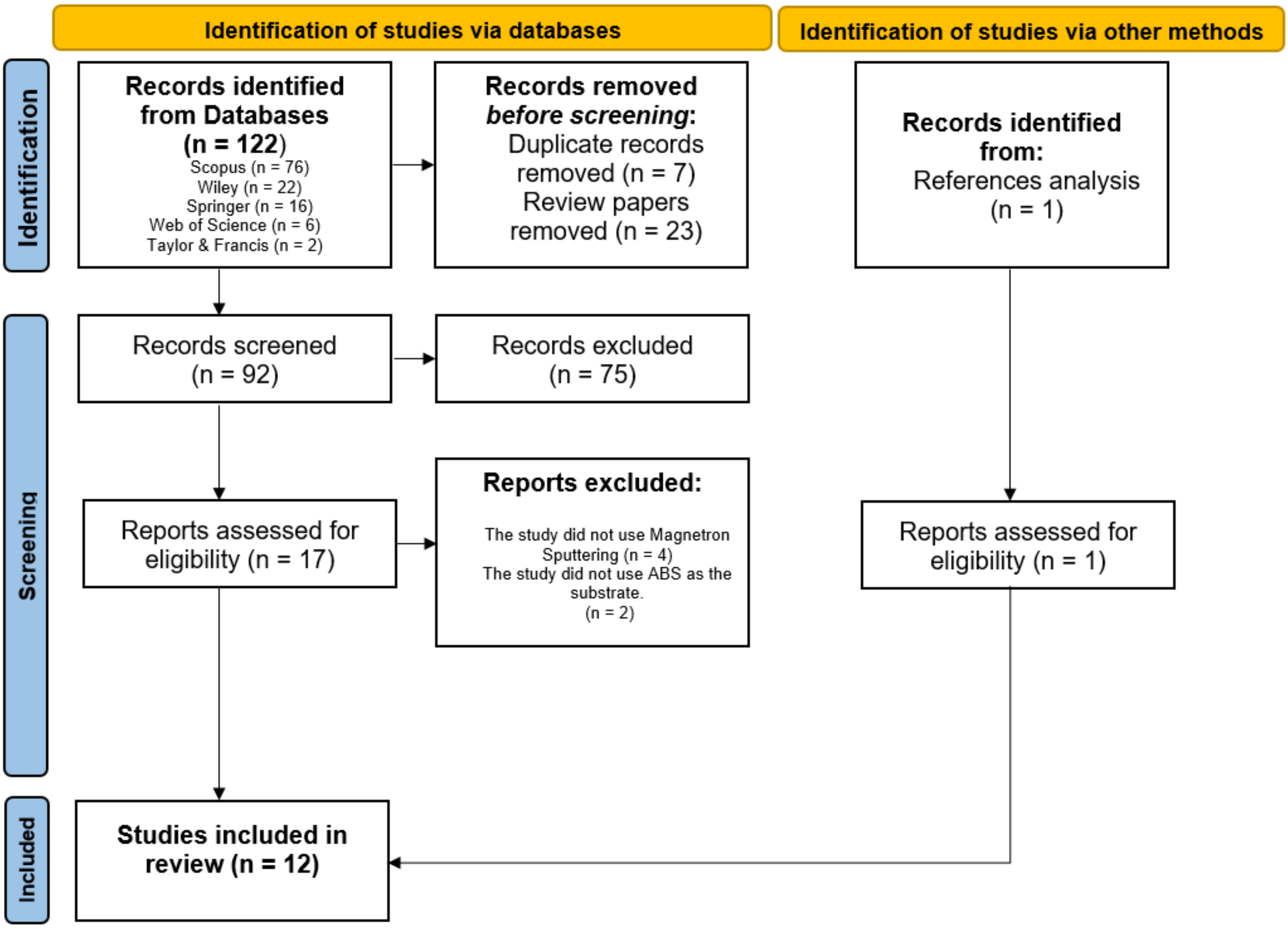

Figure 1. Flowchart of article selection steps.

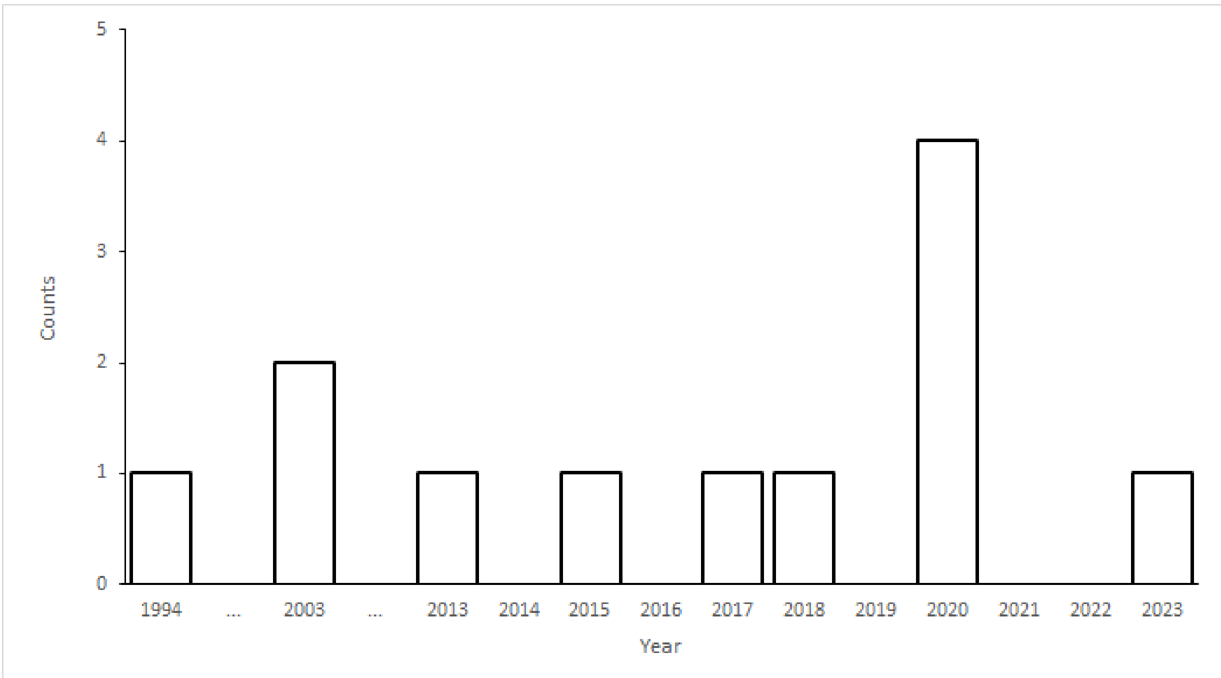

Figure 2. Number of publications per year.

Ref	Substrate	Coating	Film thickness	Pretreatment	Deposition method											
					Source	**Target**	**Target to substrate distance**	**Substrate preheating**	**Chamber pressure**	**Gas**	**Gas flow rate**	**Power**	**Sputtering pressure**	**Discharge current**	**Bias voltage**	**Sputtering time or rate**
De Bruyn *et al.* (2003)	ABS PP PC GFRP	Al	0.1 to 1 μm	Flame, plasma chemical, mechanical and lacquer	RF	Al	-	-	-	Ar	-	-	-	-	-	-
Jachowicz (2021)	ABS, PE and HDPE	Cu Al TiN TiAl	1672 nm 1684 nm 1544 nm 1430 nm	- - - -	- - - -	Cu Al Ti Ti and Al (50/50)	- - - -	- - - -	$5.0x10^{-3}$ Pa $5.0x10^{-3}$ Pa $5.0x10^{-3}$ Pa $2.5x10^{-3}$ Pa	Ar Ar Ar and N2 Ar	- - 50 sccm (N2) -	1.0 kW 1.0 kW 1.3 kW 1.0 kW	0.50 Pa 0.52 Pa 0.53 Pa 0.53 Pa	6.30 A 4.76 A 6.95 A 5.60 A	-50 V -50 V -50 V -50 V	1500 s 540 s 1800 s 2880 s
Sukwisute *et al.* (2017)	ABS	CrN	-	Ar plasma etching (10 min)	DC	Cr	7 cm	-	$2.0x10^{-5}$ Pa	Ar and N2	-	125 W 150 W 175 W 200 W	$4x10^{-3}$ Pa	-	-	120 min
Afshar and Mihut (2020)	ABS	Cu	1000 nm	-	-	Cu	-	-	$1.3x10^{-3}$ Pa	Ar	47 sccm	250 W	2.6x10-1 Pa	-	-	-
Zendehnam *et al.* (2014)	ABS	Ag	20 nm	-	-	Ag	12 cm	-	10^{-6} mbar	Ar	-	90 W	-	-	-	-
Pedrosa *et al.* (2018)	ABS PET PA	CrN	146 to 739 nm	Ar plasma (5 min, 100W)	DC	Cr	-	-	10^{-4} Pa	Ar N2	25 sccm 0 to 8 sccm		3.8x10-1 Pa 0.5 Pa	75 A/m^2	-	
Grimberg *et al.* (1994)	ABS	TiN	0.7 to 5 μm	Electroplating Cu and Ni layer and ion etching	-	Ti	3 cm	100 to 110°C		Ar N2	-	-	0.36 Pa	-	435 V	-
Zhang *et al.* (2020)	ABS	CrN	500 nm	UV painting, Argon etching	DC HIPIMS	Cr	13 cm	-	$3x10^{-3}$ Pa	Ar N2	40 sccm 25 sccm	4 kW	2.5 Pa	-	-50 V	-
Na *et al.* (2003)	ABS	TiN	200 nm	-	DC	Ti	10 cm	-	$2x10^{-6}$ Torr	Ar N2	-	200 to 500 W	20 mTorr	-	-	30 min
Lambaré *et al.* (2015)	ABS, PEEK and ABS/PC	Cu and Cu/Ti	400 nm	Ar plasma	DC and RF	Cu and Ti	13 cm	0 to 100°C	$6x10^{-5}$ Pa	Ar	-	200 W	0.5 Pa	-	0 to -300 V	10 min
Dai *et al.* (2023)	ABS	Cu	1400 nm	Plasma etching and Cr ion bombardment	DC	Cu	30 cm	-	$2x10^{-3}$ Pa	Ar	-	800 W	0.3 Pa	-	-100 V	1.5 h
Heinß and Fietzke (2020)	ABS, PC and PLA	Al	200 nm	Plasma pretreatment	DC	Al	30 cm	-	-	-	-	-	-	-	-	2 min

Table 1. Coating deposition parameters.

4. Discussion

Based on the results presented in Tab. 1, it is evident that the main strategies to improve adhesion were: surface pretreatment of the polymer; the use of intermediate layers; and modifications in the process parameters, particularly the sputtering power. The main coatings found in the literature to enhance the surface properties of ABS were CrN (Sukwisute *et al.*, 2017; Zhang *et al.*, 2020; Pedrosa *et al.*, 2018), Cu (Lambaré *et al.*, 2015; Dai *et al.*, 2023; Jachowicz, 2021; Afshar and Mihut, 2020), Cu/Ti (Lambaré *et al.*, 2015), TiN (Na *et al.*, 2003; Grimberg *et al.*, 1994; Jachowicz, 2021), Al (Jachowicz, 2021; Heinß and Fietzke, 2020; De Bruyn *et al.*, 2003), TiAl (Jachowicz, 2021) and Ag (Zendehnam *et al.*, 2014).

Activation, or functionalization, involves a temporary enhancement of a surface's chemical reactivity. The polymer surface can be activated through plasma treatment using inert or reactive gases, either as an independent process or within the PVD chamber. In the work of Sukwisute *et al.* (2017), the ABS was treated by argon (Ar) etching at a pressure of 4×10^{-3} mbar for 10 min to remove impurities. In Pedrosa *et al.* (2018), before all depositions, the substrate surfaces underwent plasma treatment for surface activation, using a 40 kHz RF generator at 100 W within an argon atmosphere (80 Pa for 5 minutes).

The aluminum coating was deposited by Magnetron Sputtering on different polymers: ABS, PC, PP, and the GFRP composite with epoxy as substrate (De Bruyn *et al.*, 2003). Several pretreatments were carried out to improve the adhesion of the coatings, which were evaluated through the cross-cut tape test (ISO 2409) and impact testing (ISO 6272). The performed pretreatments included flame treatment, chemical treatment, mechanical treatment, varnishing, and oxygen plasma treatment. The authors performed two types of plasma pretreatment. The first type was done in vacuum metallization equipment prior to deposition with RF power (1 kW, 13.56 MHz) at a pressure of 1.5 Pa O_2. The second type was carried out in a plasma treatment installation with RF (13.56 MHz) power set at 100 W and a O_2 pressure of 60 Pa. In summary, all samples showed good adhesion according to the tape test. In terms of impact testing, ABS and PC substrates exhibited higher adhesion regardless of the pretreatment.

Other studies have also used plasma pretreatment, often with the aim of surface activation. However, in general, this treatment plays an important role in sample cleaning (Zhang *et al.*, 2020; Lambaré *et al.*, 2015; Heinß and Fietzke, 2020). The application of plasma treatment to polymer surfaces can lead to surface texturing, resulting in improved adhesion strengths, primarily attributed to mechanical interlocking. Additionally, this texturing might coincide with alterations in surface chemistry, caused by changes in terminal species (Mattox, 2010).

Samples of ABS (acrylonitrile butadiene styrene) were manufactured through 3D printing for the application of copper coatings (Afshar and Mihut, 2020). After the deposition of the coatings, the samples were placed in a controlled chamber to simulate the environmental conditions, such as UV exposure and humidity, following ASTM G154 standard. Adhesion was evaluated through the cross-cut tape test and by analyzing the fracture surface after the bending test. By examining the cross-sectional image obtained through SEM, it was possible to observe that the copper film exhibited columnar growth. Despite 1200 hours of exposure to UV rays and humidity, the coatings showed excellent adhesion according to the maximum category defined by ISO 2409. Overall, the authors concluded that the PVD metallization of ABS was effective in protecting the substrate under severe environmental conditions and also improved its bending resistance.

Another method to enhance the adhesion of coatings on polymers is the usage of intermediate layers placed between the film and the substrate. The inter-layers are also known as "tie layers" or "glue layers".

Thin films deposited by PVD are known as hard coatings and, in this context, TiN stands out as an excellent coating due to its high hardness, wear resistance, and corrosion resistance. As mentioned above, adhesion and temperature are factors to be considered when a metallic film is deposited onto a polymer. In the study of (Grimberg *et al.*, 1994), elements such as structure, morphology, and adhesion of TiN coatings were investigated based on their thickness and the interlayers present between the hard coating and the substrate.

Therefore, prior to the deposition of the PVD coating, the ABS was metalized using electrodeposition, studying the following interlayers: Ni, Ni/Cu, Cu/Ni, and Ni/Cr. Then different thicknesses of the TiN film deposited on the metallized surface were investigated, ranging from 0.7 to 5 μm. SEM images revealed several cracks when depositing the TiN film directly onto the ABS, highlighting the authors' emphasis on the essential role of an intermediate layer to ensure the adhesion of the hard coating. Regarding the thickness of the coating, the authors noted that increasing the thickness led to the formation of grains with different orientations, which directly affected the properties (Grimberg *et al.*, 1994).

Titanium and Chromium are excellent materials to be used as glue layers in polymers due to their ability to form organo-metallic bonds with the substrate, improving adhesion (Mattox, 2010). In this context, Lambaré *et al.* (2015) investigated the usage of Ti, while Dai *et al.* (2023) evaluated Cr as an interlayer.

In addition to the use of tie layers, Lambaré *et al.* (2015) examined the effect of plasma pretreatments using different reactive gases, i.e. N_2 and O_2. The adhesion of the coatings was evaluated using the Scotch test (according to ISO 2409) and the pull-off test (as per ISO 4624). The authors concluded that plasma treatment enables surface functionalization, leading to a noticeable enhancement in surface wettability. This outcome was ascribed to a chemical modification of the polymer surface (resulting in the creation of polar groups on the surface), which enhances the adhesion of the metal layer.

Regarding the use of Ti as an interlayer, the results showed that, in addition to enhancing adhesion, titanium exhibited the characteristic of a diffusion barrier coating.

There are studies that employed variations in deposition parameters to enhance the adhesion of coatings deposited on ABS. Dai *et al.* (2023) combined ion bombardment techniques to improve adhesion. Recent studies highlight that sputtering power is one of the most influential parameters in thin film adhesion (Jachowicz, 2021; Sukwisute *et al.*, 2017; Zhang *et al.*, 2020; Na *et al.*, 2003).

In this way, Sukwisute *et al.* (2017) investigated how the variation in the sputtering power (from 125 to 200 W) affects the hardness and wear resistance of chromium nitride (CrN) thin films that were deposited onto ABS substrates. The CrN coating, when deposited on ABS substrate at 175 W, exhibits the highest hardness, 9.58 GPa, consequently enhancing the wear resistance of the ABS surface. Conversely, excessive sputtering power, such as 200 W, leads to structural weakening of the film due to the formation of numerous voids and cracks.

Furthermore, Lambaré *et al.* (2015) also discussed the effects of coating adhesion in relation to bias voltage and temperature. The authors observed that increasing the temperature led to an increase in the delamination stress of the coatings and poor adhesion. The authors attribute the rise in temperature to an increase in copper diffusion, which explains the property losses. Concerning bias voltage, the authors noted that increasing voltage results in a 1.3-fold increase in delamination stress.

Magnetron Sputtering has been intensely researched over the decades, and its potential to coat polymers is a current trend. Polymers benefit significantly from their ability to deposit materials at low temperatures. Nevertheless, the usage of strategies to improve adhesion is essential due to the chemical bonds in polymers. This systematic review of the literature has highlighted that pretreatments, intermediate layers, and variations in deposition parameters are effective approaches for enhancing the adhesion between the thin film and the ABS substrate.

5. CONCLUDING REMARKS

Physical vapor deposition (PVD) coatings on polymers have gained significant attention due to their potential applications in various industries, such as solar panels, smart windows, electronics, automotive, and aerospace. One critical aspect in the development of PVD coatings is the adhesion to the substrate, which directly affects the performance and durability of the coatings. This review paper examines recent studies that have focused on evaluating the adhesion of PVD coatings on Acrylonitrile Butadiene Styrene (ABS).

5.1 Coatings:

The selected studies focused on coatings such as CrN, Cu, Cu/Ti, TiN, Al, TiAl and Ag. These coatings aimed to achieve desirable properties, including conductivity, transparency, flexibility, and thermal stability, depending on the specific application.

5.2 Evaluation of Adhesion:

The adhesion of the coatings was assessed using different techniques such as risk tests, crosshatch tape tests, micro-scratch tests, and nano-scratch tests. The critical adhesion force or load (Lc) was used as a parameter to quantify the adhesion strength between the coating and the substrate.

5.3 Methods to improve Adhesion:

It was observed in the studies that the main methods to enhance the adhesion between the coating and the substrate were:

- Pretreatment: Flame, chemical, mechanical, lacquer (De Bruyn *et al.*, 2003) and plasma (De Bruyn *et al.*, 2003; Sukwisute *et al.*, 2017; Pedrosa *et al.*, 2018; Zhang *et al.*, 2020; Lambaré *et al.*, 2015; Heinß and Fietzke, 2020);

- Interlayer: Cu and Ni (Grimberg *et al.*, 1994), Ti (Lambaré *et al.*, 2015) and Cr (Dai *et al.*, 2023);

- Increasing sputtering power (Jachowicz, 2021; Sukwisute *et al.*, 2017; Zhang *et al.*, 2020; Na *et al.*, 2003; Dai *et al.*, 2023);

- Changing gas flow rate (Zhang *et al.*, 2020; Pedrosa *et al.*, 2018);

- Changing bias voltage and substrate temperature (Lambaré *et al.*, 2015).

5.4 Key Findings:

The research findings demonstrated that both the deposition power and plasma pretreatment had a significant impact on the adhesion of the coatings. Increasing the deposition power improved adhesion, while plasma pretreatment enhanced the bonding between the coating and the substrate by modifying the surface properties. The use of interlayers, such as Ti, Cu, Ni and Cr, also proved effective in enhancing the adhesion of the coatings.

In conclusion, the studies reviewed in this paper highlight the importance of optimizing the adhesion of PVD coatings on ABS. The deposition techniques, properties studied (optical, thermal, mechanical), and adhesion analysis techniques employed varied among the studies. Nonetheless, it is evident that factors such as deposition power, plasma pretreatment, and the use of interlayers play crucial roles in enhancing the adhesion of the coatings. Further research is warranted to explore additional deposition techniques and parameters that can further improve the adhesion and overall performance of these coatings.

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