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Sustainable Corrosion Protection of Aluminium Alloys – Life Cycle Assessment of Established and Innovative Coating Processes

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Abstract: Aluminium alloys are highly valued for their lightweight properties. However, their susceptibility to corrosion, particularly in chloride-containing environments, presents significant challenges especially when considering "green" sustainable processing routes. Applying ISO 14040 life cycle assessment (LCA) approach, traditional surface treatment techniques (anodization, conversion coatings, organic paints, plasma activated chemical vapour deposition in vacuum) are compared for the first time to innovative approaches such as Atmospheric Pressure Plasma Deposition (APPD). Although highly conservative assumptions are taken in the life cycle inventory, the greenhouse gas emissions of APPD are clearly lower than in the other technologies, especially because of the higher efficiency in feedstock use, the higher throughput, and the plasma-and air-supported oxidation of CH_4, formed during dissociation of the silicon organic precursor. By achieving corrosion properties of cerium-doped APPD silicone coatings similar to the state-of-the-art and elimination of repainting as for organic paints, APPD is at the forefront of sustainable processing.

Keywords: aluminium alloys; atmospheric pressure plasma deposition; coatings; corrosion protection; life cycle analysis

1 INTRODUCTION

The corrosion behaviour of aluminium alloys arises from their microstructural and surface characteristics. AlSi10Mg, when produced using conventional casting or advanced additive techniques like powder bed fusion (PBF), often has a microstructure that includes silicon particles distributed in the aluminium matrix [1]. In PBF-processed alloys, rapid solidification can lead to fine cellular or dendritic microstructures, which enhance strength but may introduce porosity and surface roughness. These microstructural features create localized galvanic cells and make the material more prone to pitting and general corrosion [2]. Moreover, the intrinsic oxide layer (Al_2O_3) that forms on aluminium provides only moderate protection and is susceptible to degradation in acidic or highly alkaline environments [3].

Traditional corrosion protection techniques for aluminium alloys include organic coatings, anodization, and conversion coatings [4]. Coatings are evaluated through standardized methods such as ISO 9227 for salt spray resistance and ISO 2409 for adhesion. Organic coatings, typically applied as paints or primers, form a physical barrier to environmental factors [5]. While providing initial corrosion resistance, organic coatings may suffer from permeability to moisture and oxygen, eventually leading to crevice corrosion at the interface to the substrate. Repainting every 5-10 years is common. Anodization as an electrochemical process generates a thick oxide layer on the aluminium surface [6]. The layer's porosity can be reduced by subsequent sealing treatments to enhance barrier properties (e.g., in hot water or nickel acetate solution). Evaluations using EN 12373 standards reveal that anodized coatings exhibit good corrosion resistance and surface hardness. However, anodization is sensitive to surface imperfections and can amplify defects, especially in components with rough surfaces. Further, acidic solutions require neutralization and hazardous waste disposal. Conversion coatings, such as chromate or phosphate coatings, provide active corrosion protection by chemically altering the substrate's surface [7]. While chromate coatings have excellent self-healing capabilities, their use is restricted due to environmental and health concerns [8]. Recent efforts have explored non-toxic alternatives like cerium-based conversion coatings [9], which show promise in mitigating pitting corrosion.

Sol-gel technology involves the application of colloidal solutions that solidify into thin films upon drying and curing for crosslinking at elevated temperatures or UV radiation [10]. Mainly silicon organic precursors are used, mixed with solvents. However, their relatively limited thickness (a few 100 nanometres due to cracking risks) and durability in extreme conditions restrict their industrial use. Similarly, plasma activated chemical vapour deposition (PACVD) relies on silicon organic precursors, being polymerized under vacuum conditions using radio-frequency, microwave or kHz pulsed plasma at the components surfaces in a thickness regime of typically 1 µm thickness [11]. Pre-treatments increase the coating adhesion, while deposition in cavities is complex.

Atmospheric pressure plasma deposition (APPD) is an innovative surface coating technology at the industrial breakthrough, overcoming main limitations of vacuum based processing of PACVD, if working with vaporized or aerosolized precursor liquids [12]. The APPD process does not require vacuum systems, making it economically and operationally attractive. Fig. 1 illustrate the setup and operation of the APPD system based on robot mountable plasma jets, highlighting its capability to process components with complex 3D geometries [13]. The admixture of organic precursors or aerosols (like polymerize-able hexamethyl disiloxane (HMDSO) with e.g. Ce containing salt or nanoparticle containing aqueous or alcoholic solutions), done directly to the atmospheric pressure plasma or to the subsequent thermal "afterglow" zone in front, leads first to fragmentation of the organic molecules as basis for cross-linking after deposition of silicones in inert or silicates in oxygen containing process gas on the substrate surface.

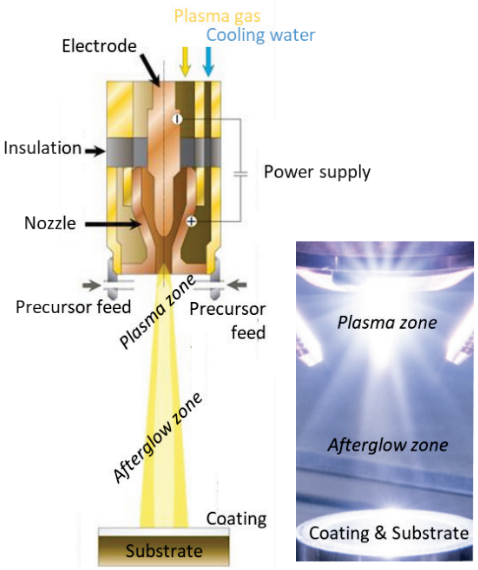

Figure 1 Principle of plasma jet based APPD for coating deposition: (left) Plasma formation by high current discharge within a cathode-anode slit at simultaneous high inert gas flow, transporting the excited species (Ar ions, electrons) to the open air in front of the jet. (right) Photograph from process control. [13]

The APPD coatings with high barrier properties are particularly effective due to their ability to incorporate active corrosion inhibitors like cerium ions from aqueous aerosols. The mechanism involves the formation of cerium hydroxides and oxides at defect sites, which suppress corrosion propagation, as shown by the excellent performance in salt spray tests of PBF components (Fig. 2a) and the dense barrier formation in impedance spectroscopy (Fig. 2b). In comparison, they are outperforming traditional coatings in both corrosion resistance and adhesion in chloride-rich environments.

The comparison of APPD with traditional methods also extends to environmental and economic aspects. APPD coatings are free of hazardous materials like hexavalent chromium, aligning with regulatory frameworks such as REACH and RoHS. The process's energy efficiency and scalability further enhance its industrial feasibility, particularly for applications requiring high-throughput and cost-effective solutions, which is shown in the following by an extended LCA in detailed comparison to state-of-the-art PACVD in vacuum and using literature data to conventional processes.

2 EXPERIMENTAL

The environmental sustainability of coating technologies can be evaluated systematically by LCAs using standardized methodology (e.g. ISO 14040), assessing the environmental impacts of a product or process over its entire lifecycle from raw material extraction to end-of-life disposal. Based on own data on the typical annual performance of a APPD plant for deposition of 1 µm thick coatings in 1 shift operation, 7200 m²/year coating area were defined, i.e. for a 10 years equipment use 72,000 m² coated surface. The deposition of coatings with comparable properties by PACVD requires ~3.8 units because of the longer process times. Salt spray testing of rough PBF AlSi10Mg components revealed, that the specified minimum lifetime is fulfilled by such 1 µm coating thickness, being basis for the comparative assumption for the conventional technologies.

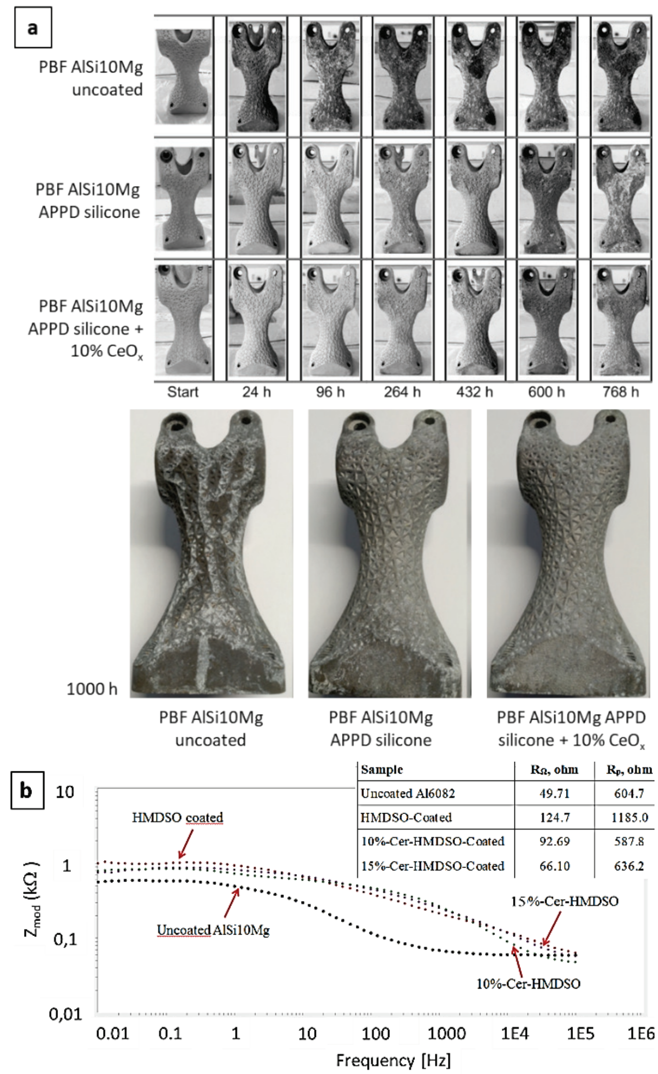

Figure 2 (a) PBF AlSi10Mg components with rough, ball-blasted surfaces after up to 1000 h salt spray testing (ISO 9227) (b) Active corrosion protection effect (barrier effect & healing of defect sites) of APPD silicone coatings revealed by impedance spectroscopy measurements. [13]

The Life Cycle Inventory (LCI) on all inputs (materials, energy) and outputs (waste, emissions) throughout the lifecycle stages is given in Tab. 1, laying the focus especially on the initial processing and excluding the end-of-life of the coated component (i.e. due to negligible effects of such silicone coatings in metal recycling) and of equipment (cradle-to-gate approach).

The analysed impacts include greenhouse gas emissions, resource depletion (fossil and minerals/metals), water consumption, taking the impact factors from the Environmental Footprint method. Resource depletion-fossil and water consumption follow the same trend as GHG emissions and are thus not reported. Background data have been taken from ecoinvent v3.10 database [14].

Table 1 LCI for silicone coating processes and equipment – APPD vs. PACVD

Parameter	APPD	PACVD
Functional Unit (FU)	1 µm silicone on 7,200 m² (annually) and 72,000 m² (equipment life)	
Annual Energy Use (MWh)	144	2,080
Gas Consumption (m³/a)	18,000 Ar	1,043 N_2
HMDSO Feedstock Use Efficiency (%)	60	50
Totally required HMDSO Precursor (kg/a)	216	432
Methane Emissions (CH_4, kg/a)	42,7	85,3
Equipment Lifespan (years)	10	10
Number of required Units (based on achievable deposition rate to enable 7200 m²/a)	1	~3,8
Metals used per Unit – Steel / Stainless Steel / Copper / Brass / Aluminium / Titanium (kg)	1314 / 74 / 81 / 6 / 288 / 0	115 / 460 / 86 / 6 / 115 / 6
Polymers / Electronics used per Unit (kg)	79 / 34	63 / 259
Maintenance Components per Year	30 plasma nozzles, 5 coatings	Protective shields (1 per year), pumps (1 each 4 years)

3 RESULTS & DISCUSSION

The assessment of potential environmental impacts targets as main indicator the greenhouse gas emissions (GHG), for which both the equipment manufacturing (raw materials and device manufacturing) and the coating deposition itself is taken under consideration; excluded are emissions for aluminium substrate components are not considered, being similar for the envisaged coating processes. Tab. 2 shows the accurate calculation results for APPD and PACVD and published average values for the state-of-the-art technologies [15-21].

Table 2 Energy requirements and emissions for coating technologies for corrosion protection of aluminium [12-18]

Parameter	APPD	PACVD	Anodization	Organic Coatings	Conversion Coatings	Sol-Gel Coatings
Annual Energy Use (MWh) for 7200 m²	144	2,080	400	50	120	100
CH_4 Emissions (kg/year)	42.7	85.3	Negligible for the surfacte modification step (i.e., excluding the production of feedstock)			
GHG Emissions (kg CO_2e/m^2)	1–2	6–8	5–7 (with sealing)	8–12	2–3 (cerium-based)	1–2
Material Efficiency (%)	60	50	Moderate	Low	High	Moderate
Waste Generation	Low	Moderate	Moderate	High	Low	Low

Based on the precise insight in PACVD and APPD processes, GHG as well as the abiotic depletion potential (ADP) can be well split to the contributions from the coating device (including an annual depreciation of construction materials and manufacturing processes for the device), from necessary spare parts in maintenance, and from the processing step (i.e. the coating application and the contributions from required HMDSO precursor and gas feedstock) (Fig. 3).

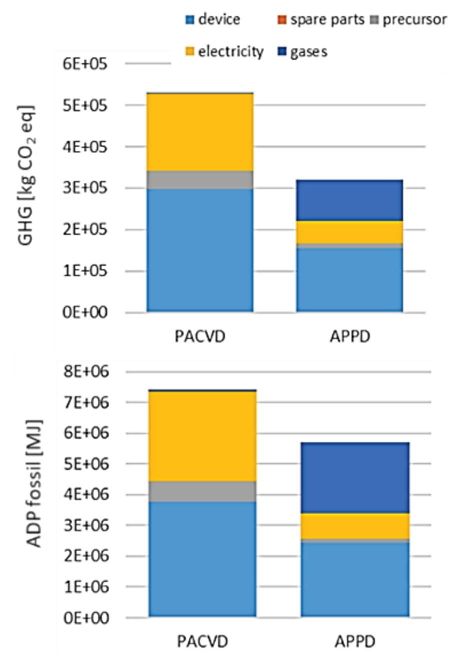

Figure 3 Comparison of GHG (above, CO_2 eq) and ADP (below) for PACVD and APPD, related to a functional unit ("coating device")

Well visible is the high contribution of the devices and the electricity necessary for PACVD processing as well as the high GHG contribution from the precursors. Importantly, the given data is shown per functional unit, which means for comparable annual coating area of 7200 m² 3.8 PACVD units need to be applied, while a single APPD unit is sufficient. In contrast, the construction of one APPD device is much less demanding in terms of materials. Further, the lower electricity consumption during processing as well as the minimization of CH_4 formation as exhaust gas, due the precursor molecules leaving the plasma reduce significantly the GHG emissions. Additionally, the efficiency in material use is at least 10% higher for APPD than for PACVD (50 vs. 60%).

Having a closer look on the contributions of the various materials used for the device construction, some significant differences are visible (Fig. 4). PACVD vacuum coating demands a heavy stainless steel vacuum chamber and a massive carbon steel frame, while constructions for APPD are based on carbon steel for the frame too, but the deposition enclosure to prevent exhaust gas emission directly to the operators and the used manipulation robots are mainly based on aluminium.

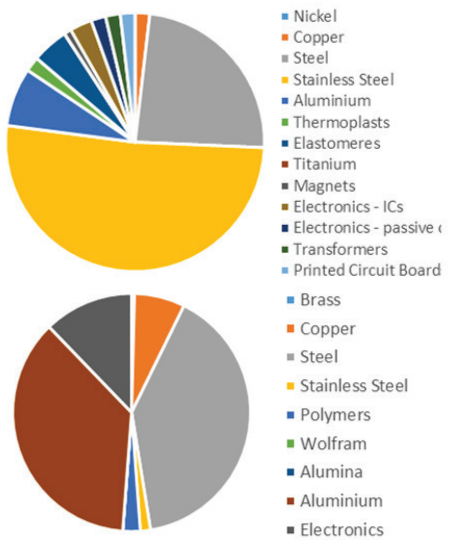

Figure 4 Comparison of GHG (CO_2 eq) on the materials used for construction PACVD (above, ~297,000 kg CO_2 eq in total) and APPD (below, ~156,000 kg CO_2 eq) for industrially-scaled coating deposition

Comparing the results with state-of-the-art processes (Tab. 2), based on literature provided data [15-21], the reasons for the observed differences of conventional processes and PACVD to APPD are exemplary in terms of

- energy requirements: demands of heating for anodization (incl. sealing) and drying / curing of organic paints
- GHG emissions; during device manufacturing and processing
- material utilization and waste: localized APPD deposition with very low overspray compared to typically 20-30% overspray in compressed-air spraying of organic and sol-gel coatings
- material footprint; for the construction of tools
- by-products and emissions: acidic and metallic effluents from solvents in organic and sol-gel coatings during sealing (incl. potentially hazardous nickel acetate)
- process complexity and scalability: extensive surface pre-treatment in anodization for uniform defect-free oxide layer formation, and a strong environmental and humidity impacts on sol-gel curing.

4 CONCLUSIONS

Corrosion protection of aluminium alloys is crucial for their long service-life, but a quite complex task with sustainable technologies with no environmental harms. As shown by first time LCAs for protective silicone coatings, the innovative APPD processes combines high ecologic potentials compared to state-of-the-art vacuum PACVD and other established processes from literature. Based on salt spray test results, the demanded service life is easily achieved by high-performance, high-throughput APPD treatments. Comparatively, PACVD faces limitation in scalability due to required vacuum conditions, while conversion coatings, anodization and organic coatings require enhancements to meet modern sustainability standards by emissions, durability and hazards.

Nevertheless, the detailed view on the device construction and use for silicone, Cerium doped corrosion protective coating deposition provides also insight into potential areas for further decrease of GHG emissions: Rethinking device construction to minimize the impacts from the machinery hardware and redesign processes to reduce Argon as process gas, either by gas recycling or use of less clean gas, can be drawn for future development steps.

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