

Interface and volume transport on technical cathaphoretic painting: A comparison of steel, hot-dip galvanised steel and aluminium alloy



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ABSTRACT

Water diffusion through the polymer and the ion transport along the coating-metal interface is examined on steel, hot-dip galvanised steel (HDG) and AlMg(Mn) with clear and pigment cathaphoretic paintings (E-coats). A general velocity decrease is identified from AlMg(Mn) to HDG steel to steel via extensive impedance studies combined with force modulation analyses. The polymer matrix reveals a slight *Young's modulus drop* and an increased disorder in the pigment-polymer network on rough substrates after 50 days of salt-water uptake. Further signs of plasticising are not observed but these circumstances already require a *Non-Fickian approach*. During the film formation, the substrate dissolves due to the extreme cathodic polarisation under high alkaline conditions, which is why the E-coat is contaminated by the solvated metal on zinc and aluminium and – contrary to expectations – with its highest contamination on steel. The *Kelvin Probe data* illustrates the prominent differences between the clear and the pigment E-coat interfaces as well as the concentration- und substrate-effects under humid oxygen-deficient conditions. Additionally, the interfacial bonding properties of the E-coat with aluminium oxide are used to assess the pH-activities during the sub-surface flux to complete this comprehensive study about the substrate dependencies.

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1. Introduction

Diffusion of corrosive species through polymer coatings and the transport along their metal interfaces plays a fundamental role in the lifetime of coating-metal systems [1,2]. The polymer permeability for water molecules and solvated ions causes a swelling, in which water acts as a plasticiser. The chemical properties of the coating change and the polymer network passes on to an amorphous state. The salt-water transport occurs at the polymer-metal interface either after the water penetrates through the whole polymer or due to substrate deep defects in the coating. Solvated ions like chloride or sulphate increase the conductivity, which increases the corrosion rate. Combined with a humid oxygen atmosphere, these conditions are the perfect environment for cathodic delamination [3], filiform corrosion [4,5], blistering [6] and, lastly, the complete deterioration of the coating.

Especially water-borne cathaphoretic paintings (E-coats) – the first protective shield of automobile bodies – have the major

function of resisting corrosion and de-adhesion. Commonly, a conversion of an epoxy resin with amino alcohol forms a polyamine with hydroxyl groups. The resulting resin is cross-linked by a blocked aromatic isocyanate and forms a polyurethane network. During electrophoresis, the positively charged amine is neutralised to the metal cathode and therefore the resin is deposited homogeneously on the metal substrate [1]. In spite of the excellent corrosion resistance of these paintings, de-adhesion on steel and zinc substrates in the form of blistering is observed along the interface and far away from any defects. By contrast, anodic undermining dominates on the boundaries of aluminium and low-alloyed steel [2]. The complex interactions at the different polymer-metal interfaces, coupled with the plasticising of the pure coating, hinder a simple explanation of the practical corrosion and infiltration phenomena.

Actual research is focused on typical failure by corrosion with assumed Fickian swelling in an oxygen atmosphere. The barrier coating properties are modelled via electrochemical impedance spectroscopy (EIS), which characterises the failure due to interstitial stagnant moisture [7]. Unfortunately, only a few EIS studies consider the transition from Fickian to Non-Fickian behaviour during water uptake as summarised in Fig. 1. First,

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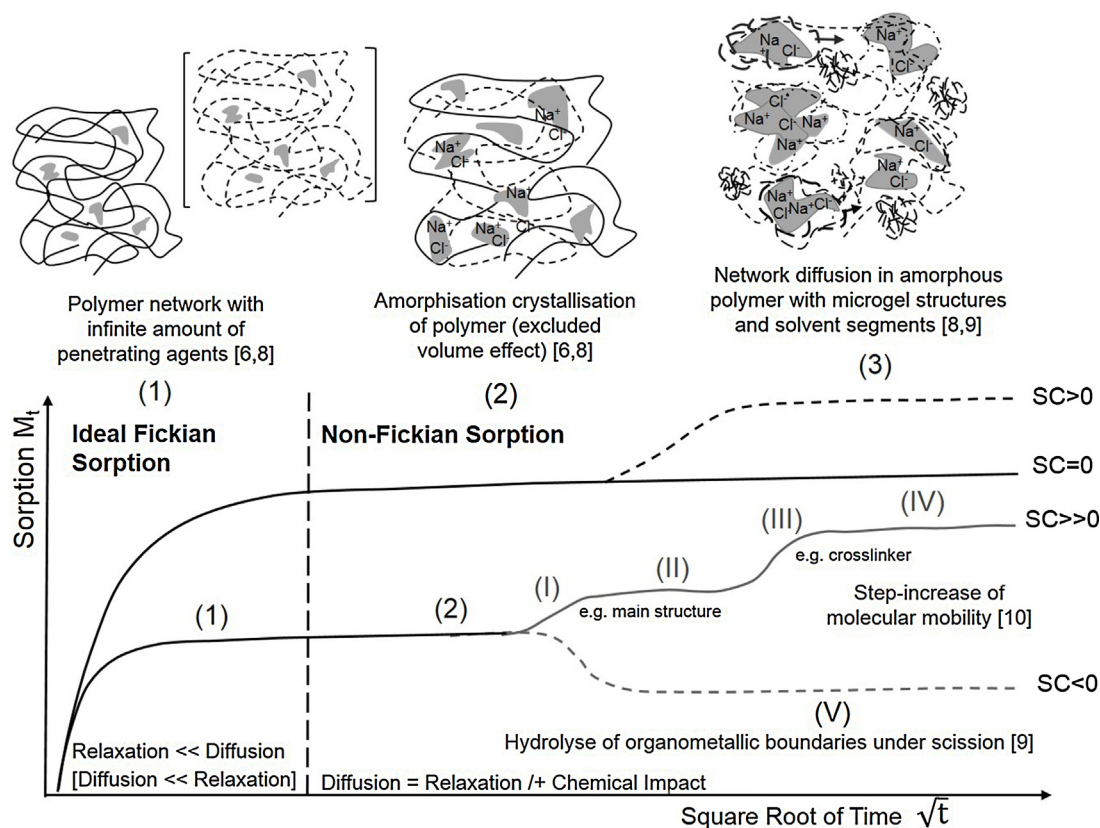


Fig. 1. Review of the polymer mechanism (top) and the diffusion kinetic (bottom) during the moisture swelling referred to [6,8–10].

the relaxation or the diffusion is rate-determined (1), then, the amorphisation crystallisation of the polymer (2) starts and the transport processes get disordered [6,8–10]. Dependent on the coating, microgel structures and solvent segments diffuse in networks [8] and/or the molecular mobility increases in steps on account of the different interactions of the backbone and crosslinkers to water molecules [10]. Scission is also possible due to hydrolysing of organometallic boundaries in the presence of titanium dioxide pigments [9]. With regard to stone-chipping defects, time-dependent Volta potential changes – measurable via scanning Kelvin Probe (SKP) – describe the failure progress along the polymer-metal interface. Anodic and cathodic undermining is intensively studied on model coatings excluding functional additives, pigments or fillers on steel [11–15], electro-galvanised steel [16–18] and aluminium alloys [19–21]. Indeed, transport quantification data is rare and a comparative study about the substrate dependencies, especially on technical coatings including pigments, has not been available until now. Due to the omnipresence of technical coatings, robust studies about de-adhesion of these coatings are necessary.

This work will point out the substrate-dependent water and ion transport with a focus on the polymer-pigment bulk and the polymer-metal interfaces by quantifying the transport rates. The peculiarities of technical clear and pigment E-coats were characterised by infrared spectroscopy in attenuated reflection (IR-ATR) and energy dispersive X-ray (EDX) on profile cuts via focused ion beams (FIB). Current density-potential curves are used to describe the substrate behaviour of steel, hot-dip galvanised (HDG) steel and aluminium alloy AlMg(Mn) in various pH buffers. The water diffusion through the coating volume is quantified via Fickian and Non-Fickian approaches using extensive EIS data. Independent resistance and confidence analyses support the

diffusion determination with the applied circuit. The level of polymer plasticising and the roughness changes are described via Force modulation microscopy (FMM) to identify the Fickian state after salt-water uptake. The SKP is used to compare the interfacial ion transport at the clear and pigment E-coat in a humid nitrogen atmosphere. Further, the metal-dependent kinetic data was assessed on the pigment E-coat, with 0.1 M, 0.5 M and 1 M NaCl defects. A resulting theory about the pH-activities along the aluminium interface is given and supported by confocal Raman spectra and EDX mappings. In brief, the results clarify the general differences between technical clear and pigment coating-metal systems, as well as the substrate-dependent circumstances for later corrosion and de-adhesion due to electrolyte ingress on an E-coat.

2. Experimental

2.1. Metal preparation and coating application

Steel, HDG steel and AlMg(Mn) samples (Gardabond E HDG, C and F (5005A), Chemetall GmbH) were cleaned by soaking them in toluene, acetone and ethanol, followed by submerging them in alkaline cleaner Gardoclean[®] 855/1 (20 g/L, Chemetall GmbH) and stirring them at 500 rpm at 60 °C for 5 min. Then, the samples were soaked and jetted with distilled water and dried in recirculated air at 40 °C for 120 min. The clear and pigment E-coats (Aqua EC3000, Axalta Coating Systems Germany GmbH) were deposited at 32 °C and with an initial voltage of 260 V (Wächter GLT 400/20), the metal substrate served as a counter electrode. Film thicknesses of 20 $\mu\text{m} \pm 1.5 \mu\text{m}$ were deposited on steel, HDG steel and AlMg(Mn) and were measured with PHYNIX[®] Surfifx after hardening at 180 °C for 20 min.

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