Surface modification of Cr(III) packaging substrates for enhanced adhesion via citric acid processing

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Abstract

Changes in legislation have led to Cr (VI) being removed from manufacturing processes, something that has an effect on the production of chromium coated steel products, such as Electro Chromium Coated Steel (ECCS). Previous work has shown that a novel substrate, produced using Cr(III) in the electroplating process, requires optimisation and development to give equivalent performance to the ECCS substrate for lacquered packaging applications. The passivation and pre-treatment of stainless steel has been widely investigated through both nitric acid and citric acid, showing that citric acid is a viable alternative for promoting the dissolution of anodic surface contaminant. As part of the development of a Cr(III) plated steel substrate, shown here is work investigating the use of citric acid as a pre-treatment for the developmental substrate, to improve adhesion performance of a lacquer through the sterilisation process. Through optimising the conditions by which pre-treatment in citric acid is conducted, the adhesion quality of an organic lacquer can be substantially improved, owed to the changed in chemical and physical surface characteristics induced by the process.

Keywords: Steel packaging; Cr(III) electroplating; passivation; enhanced adhesion; corrosion protection.

Nomenclature: Electrolytically Chromium Coated Steel - ECCS, Bisphenol-A non-intent – BPANI, Epoxyphenolic – EP

1. Introduction

Metal packaging is an important means by which food can be stored in modern society, resulting in a global market of around $110 Bn in 2017[1,2]. Steel based metal packaging boasts many benefits, in that it is robust, recyclable, achieves many years of shelf life, reducing waste for both producer and consumer, and has established products with whole supply chain integrity[3,4]. Traditionally, steel packaging has been produced either using tinplate, having a layer of tin approximately 0.2-2µm thick or electro chromium coated steel (ECCS) which has a 10-30 nm layer of chromium/chromium oxide, produced using a Cr(VI) electroplating process[5].

The steel packaging industry, as with other European industries, faces the challenge of removing Cr(VI) from industrial processes due to legislative moves, driven by health implications surrounding Cr(VI) species[6]. To this end, novel methods to electroplate steel with chromium species have be developed, using Cr(III) species in the electroplating process[7], which replace the Cr(VI) species used in the production of ECCS. The novel substrate is one that requires the same level of characterisation as the well-established ECCS. To this end, previous work carried out using iterations of this novel developmental Cr(III)

derived substrate showed that it is less resistant to corrosion when compared to the Cr(VI) derived substrate[8].

The primary usage for this substrate is to be for coated applications where an organic lacquer is applied and cured prior to formation and filling of a packaging can. The coating provides a protective layer during handling, can forming, filling and cooking processes. Bisphenol-A (BPA) has been a key monomer in the formation for epoxyphenolic lacquers, providing a flexible and versatile coating. However, owing to legislative moves in some European territories, BPA must be removed from food contact materials [9]. Coating chemistry technologies based around polyester (BPANI - BPA non-intent) have been developed [10,11], which have been found to be perform poorly compared to the incumbent epoxy phenolic materials, [12][13].

Following filling, the can contents is cooked and sterilized under temperature and pressure. Under this aggressive environment the novel, developmental Cr(III) substrate performs less favourably than ECCS [14]. When the substrate is subjected to the sterilisation process, corrosion is induced at the surface of the substrate, owing to the aggressive chemical environment that is contained within the can at high temperature and pressure [15] which leads to failure at the lacquer / substrate interface. There is significant interaction between the foodstuff (through controlled simulant) and the substrate/coating system, showing the novel Cr(III) substrate is a promising alternative to ECCS but with development required to show equivalent performance[14]. A feature of this previous work is the advantage of using citric acid in the sterilisation process, seemingly mitigating the effects of sodium chloride[12,14] in improving the lacquer / substrate adhesion. This was attributed to an increase in chromium oxide species on the surface and a reduction of iron species compared with the pre-sterilisation surface chemistry.

Citric acid has been examined as a pre-treatment or passivation technique for stainless steel[16], as a replacement for nitric acid. The purpose of the passivation technique is to remove anodic surface contaminants such as iron compounds from the surface through chemical dissolution, leading to the formation of an oxide layer[17], hence improving corrosion resistance, an important property in many industries, including the steel packaging industry. Citric acid is interesting due to it being present in the juices of citrus fruits. Due to its biological source and biodegradable nature, it is a more sustainable and less toxic alternative to nitric acid.

There is a potential benefit of applying a citric acid based pre-treatment on the novel Cr(III) plated steel substrate during manufacturing in order to enhance the substrate / lacquer interface integrity. The aim of the investigation is three-fold. Firstly, it aimed to understand the mechanism by which the adhesion of an organic lacquer is improved by a citric acid based pre-treatment. Secondly the study aimed to identify the interrelationship between the citric acid pre-treatment and other foodstuff simulants. Finally, it aimed to assess the feasibility of applying a citric acid pre-treatment for Cr(III) coated steel packaging substrates. This was achieved by studying surface chemistry, the electrochemical and corrosion properties, topological properties as well as the macro scale adhesion properties that have the greatest impact for the can manufacturers and fillers.

2. Materials and Methodology

2.1 Materials

Cr(III) electroplated substrate was supplied as a developmental substrate from TATA steel packaging, using Cr(III) in the electroplating bath rather than the Cr(VI) species used in ECCS production. The developmental substrate was produced on a full-scale electroplating line during parameter optimisation tests and is hence not commercially available. ECCS used as a reference material was also supplied by TATA. Neither substrate has free Cr(III) or Cr(VI) present on the surface after the electroplating process, chromium then being present in the form of chromium metal, oxide or hydroxide.

The coatings used for sterilisation testing were an epoxy-phenolic (EP) lacquer and a polyester Bisphenol-A (BPA) non intent (BPANI) lacquer, both of which were supplied by Metlac. For the purposes of this research, substrate samples were coated with 10-12 μm of either the EP or BPANI coating, cured in accordance with product guidelines, then cut to 120 mm x 45 mm for sterilisation and adhesion testing.

2.2 Citric Acid Pre-treatment

Samples of the Cr(III) electroplated substrate were submerged in a solution of 0.104 mol dm^{-3} (2% w/v) or 0.2603 mol dm^{-3} (5% w/v) citric acid, held at a temperature of either 90°C or 95°C for varying amounts of time. The solution was stirred at a constant rate to allow the diffusion of the citric acid species to the surface of the metal sample.

2.2 Sterilisation

To simulate the sterilisation process, the coated substrate samples were places in sealed jars, filled to 80% of total capacity with simulant solution. The filled and sealed jars were placed into a CertoClav MultiControl 2 autoclave and subjected to sterilisation for 90 minutes at 121°C.

2.3 Adhesion Measurements

Adhesion measurements were conducted three ways. Firstly, the adhesion failure force was measured via scratch testing, carried out in accordance with ISO 1518-1:2011[18]. This method provides a quantitative and repeatable analysis of the failure force of a given coating/substrate system. In addition to this, cross hatch testing was carried out in accordance with ISO 2409:2013 [19], following a technique used widely in industry for rapid assessment of adhesion. For reference, viusal inspection was also used to observe the surface and scored next standard images of failed adhesion with 5 being good and 1 being bad.

2.4 ToF-SIMS

Time of Flight Secondary Ion Mass Spectrometry (ToF-SIMS) analysis was conducted on a Scientific Analysis Instruments (SAI) MiniSIMS-ToF, using a gallium source to obtain secondary ion spectra in the form of images, giving elemental maps of the surface of the substrate. The images were obtained and analysed using MiniSIMS software.

2.5 XPS

X-ray Photoelectron Spectroscopy was used to obtain quantification of the surface species present after pre-treatment. Measurements were taken using an Axis Supra XPS system using a monochromated Al Kα source and large area slot mode detector using a 300 µm x 800 µm analysis area. Binding energies were calibrated to the main hydrocarbon peak (284.8 eV). Spectra were subsequently analysed in CASA software with Shirley backgrounds.

2.6 Scanning Vibrating Electrode Technique

SVET was used to measure the local current density distribution across the sample surface. Operation, design and calibration of the SVET instrument have been described elsewhere[20,21]. 50 mm x 50 mm samples were degreased using ethanol and a 10 mm x 10 mm central square left exposed to allow data corrosion data collection in the area. Samples were immersed in a 0.171 mol dm^{-3} NaCl electrolyte bath at 20 °C. The SVET microtip was held at a fixed height of 100 µm above the metal surface. The submerged microtip was scanned over the surface of the test samples immediately from the point of initiation and at one-hour intervals, for 24 hours. Data collected was processed to add the calibration factor to each point of the scan. Time-lapse 3D colour maps of current density were created using Surfer^{TM} from Golden Software for each scan, detailing the formation of anodes and cathodes across the metal surface.

2.7 Atomic Force Microscopy (AFM)

The sample topography was measured using a JPK Nanowizard 3 atomic force microscope (AFM). Samples were cut into 1 x 1 cm squares and mounted on a glass slide. The AFM was operated in alternating contact (AC) mode whereby the tip was vibrated at a frequency of 75 Hz and a scan rate of 0.7 line/s. The advantages of using AC mode is to eliminate the lateral forces, which causes less damage to the sample surface. Images were produced at a scan size of 15 x 15 µm. Gwyddion, a data analysis software was used to process the images and to determine average roughness (Ra) and the standard deviation of the roughness profile (Rq).

3. Results

The strategy employed in the study was to firstly explore the interaction between the presence of citric acid as a passivation treatment in the simulant solution and the presence of increasing levels of NaCl. This would identify the envelope of passivation that could be obtained within the sterilisation process. These findings then guided the study of the effectiveness of citric acid pre-treatment and its operational process window which was established by macro adhesion measurements coupled with investigative analytical techniques in order to identify key mechanisms.

3.1 Lacquer adhesion following sterilisation

Following previous work[12,14], initial experiments examined the effects that adding varying amounts of citric acid to the food packing simulant would have on the adhesion quality of the coating/substrate system post-sterilisation. The results of these experiments, obtained through scratch adhesion testing, are displayed in Table 1 and

Table 2. In each table, the cells represent the good (green) to bad (red adhesion failure forces. Typically 1N – 4N failure force represents a failure of the coating/substrate interface, failure forces greater than 9N represent an adequate level of adhesion for this application.

The performance of the epoxyphenolic lacquer is higher than that observed of the BPANI coating, which is in line with previous literature[12,14] and can be attributed to the improved barrier and adhesive properties of the epoxyphenolic coating. The net positive impact of the citric acid addition in the sterilisation process is evident from the first column of both Table 1 and

Table 2.

There is a complex interaction of the NaCl and citric acid concentration. Without the presence of the citric acid, the presence of any NaCl in the packing simulant has a dramatic impact on the measured adhesion. At higher NaCl concentrations, increasing the citric acid concentration improves the adhesion, although it does not allow the adhesion to achieve zero NaCl levels. At each NaCl concentration there is a citric acid concentration at which optimum adhesion is achieved, depicted as light green cells along the leading diagonal of the array. Beyond this optimum value, increasing citric acid concentration has a detrimental impact on adhesion.

It is postulated here that the relationship between the concentrations is due to the oxygen scavenging properties of the citric acid [22], likely to be mitigating the corrosive effects of the NaCl in solution during the sterilisation process. This would also correlate with the adhesion performance observed in previous work where deaerated testing has shown an improvement in adhesion quality when oxygen is removed [14]. Also based on previous work, it is plausible that the surface composition of the substrate is being altered by the acidic conditions, in this case, iron dissolution is likely to be being promoted.

Table 1: Cr(III) Plated substrate/BPANI coating system adhesion failure values with varying NaCl and Citric Acid concentration.

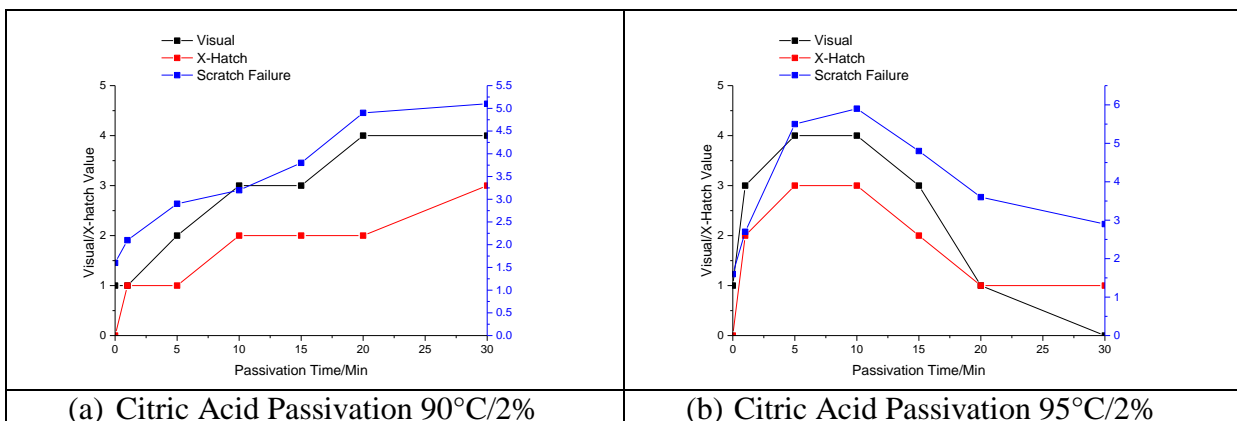|			NaCl Concentration						
	mol/L		0	0.043	0.086	0.128	0.171	0.342	0.856
	mol/L	% (w/v)	0	0.25	0.5	0.75	1	2	5
Citric Acid (mol/L)	0	0	13.1	2.5	1.5	1.0	1.0	1.0	1.0
	0.013	0.25	9.8	8.3	4.9	4.4	4.2	3.4	2.0
	0.026	0.5	10.2	6.4	7.6	5.9	5.1	4.4	2.9
	0.039	0.75	12.3	4.9	6.4	8.6	5.4	4.7	3.4
	0.052	1	14.7	3.9	4.4	6.9	7.4	6.1	3.9
	0.104	2	16.8	3.4	4.2	6.1	6.4	7.6	5.4
	0.260	5	16.8	2.9	3.9	4.9	5.6	6.9	7.1

Table 2: Cr(III) plated substrate/EP coating system adhesion failure values with varying NaCl and citric acid concentration.

		NaCl Concentration						
	mol/L	0	0.043	0.086	0.128	0.171	0.342	0.856
mol/L	% (w/v)	0	0.25	0.5	0.75	1	2	5
0	0	12.3	4.1	3.9	3.5	2.4	2.3	2.0
0.013	0.25	10.1	9.3	6.9	5.6	5.4	5.1	2.9
0.026	0.5	13.2	8.8	9.6	6.4	5.9	5.6	3.4
0.039	0.75	13.8	8.6	8.3	8.7	6.4	5.9	4.7
0.052	1	14.6	7.4	7.8	6.4	7.8	6.9	4.9
0.104	2	14.9	6.9	7.4	5.1	6.9	7.4	5.1
0.260	5	16.8	5.6	5.1	3.9	4.9	5.9	6.4

(Left axis label: Citric Acid (mol/L))

As there was a clear benefit of the presence of the citric acid on the substrate during the sterilisation process (at the corresponding NaCl conditions), it was decided that its use as a pre-treatment for the Cr(III) plated substrate should be investigated as a method by which the adhesion of an organic lacquer could be improved.

3.2 Adhesion with Citric Acid Pre-treatment

Samples of the Cr(III) substrate were placed in a bath of preheated citric acid solution at 2% w/v and 5% w/v concentration at 90°C or 95°C. These concentrations and temperatures were chosen as preliminary testing had shown them to be the optimum conditions, lower concentrations and temperatures were shown to have negligible impact within reasonable timescales. Samples were removed from the citric acid bath at regular intervals, lacquered with BPA-non intent polyester coating and subjected to a sterilisation test with a 1%NaCl solution, thus experiencing one of the harshest sterilisation environments from Table 1. In all instances there is a good correlation between the adhesion measurement methods used, Figure 1. At the lowest concentration and temperature, increasing the residence time improves the measured adhesion. As the concentration or temperature increases, an optimum residence time is achieved where further exposure to the citric acid has a detrimental impact on the adhesion. The exposure time to achieve the optimum value reduces with increasing citric acid concentration and temperature. As with the sterilisation results in Table 1 and 2, this suggests that the presence of the citric initially may have a beneficial impact on adhesion, but that excess exposure has a detrimental impact.

| (a) Citric Acid Passivation 90°C/2% | (b) Citric Acid Passivation 95°C/2% |

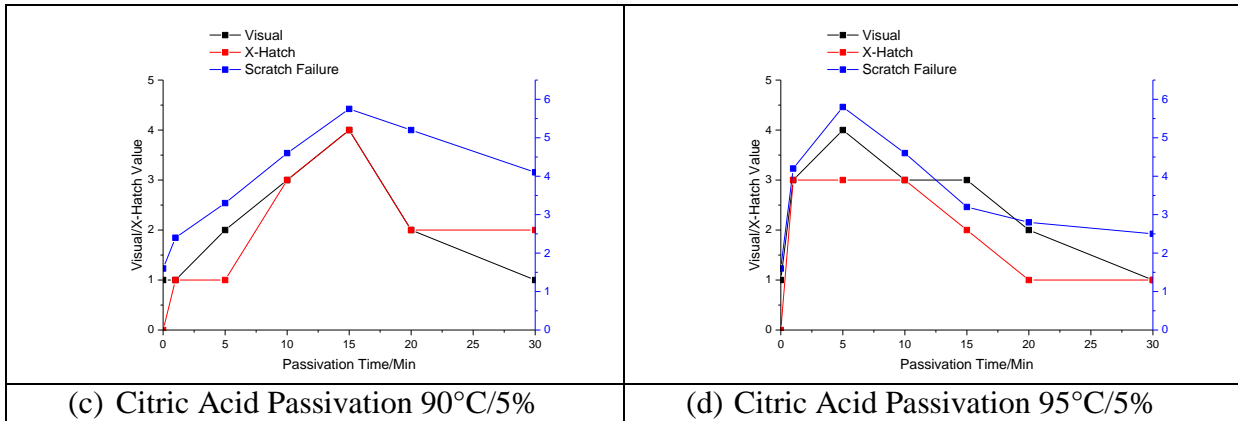| (c) Citric Acid Passivation 90°C/5% | (d) Citric Acid Passivation 95°C/5% |

Figure 1: Measured performance (quantified by scratch, cross hatch and visual appearance) of the Cr(III) substrate when subjected to citric acid pre-treatment.

As the trends observed were the same regardless of the processing parameters, for further quantification, samples for further quantification were subjected to pre-treatment at 95°C with a concentration of 5% citric acid as production rates would dictate the lowest possible exposure time in order to maintain productivity.

3.3 Surface Chemistry Characterisation

The impact of the citric acid pre-treatment on the surface chemistry of the Cr(III) plated substrate has been studied using ToF-SIMS and XPS. ToF-SIMS analysis was used to establish the surface elements and species. Images generated show the coverage of Cr (Green), Fe (Blue) and O/OH (Black) species on the surface, Figure 2.

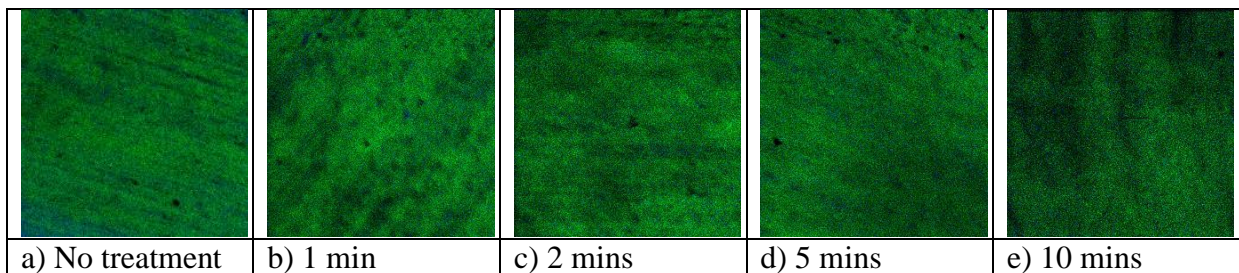| a) No treatment | b) 1 min | c) 2 mins | d) 5 mins | e) 10 mins |

Figure 2: ToF-SIMS images of the surface post pre-treatment. Cr (Green), Fe (Blue) and O/OH (Black) @ 4%, 95 °C for at each exposure time.

The effect of the citric acid exposure can be explained further by examination of the surface species at each time interval. During the initial surface exposure to citric acid (up to 5 minutes), there is an increase in the chrome oxide Figure 3(a & b) , and a complimentary decrease in chrome hydroxide species (Figure 3(c)) and chromium metal, Figure 3(d) indicating surface oxidation. During the first 5 minutes, there is a reduction in iron (Figure 3 (e)) and an increase in iron oxide (Figure 3 (f)) again indicating oxidation. After 5 minutes (the point of maximum adhesion), there is reduction in the chrome oxide and a slight increase in the chrome metal. An increase in both surface iron and iron oxide is also observed after 5 minutes.

The proposed mechanism for the changes observed is that beyond 5 minutes there is penetration of the chrome layer exposing the surface under layer of iron. The increase in

chromium metal is associated with the exposure of further the metallic subsurface beneath the surface oxide layer. The optimum adhesion conditions are therefore produced when the citric acid has produced a chromium oxide rich surface, surface iron species are removed but there has been no penetration through the metallic chromium layer which exposes the iron subsurface.
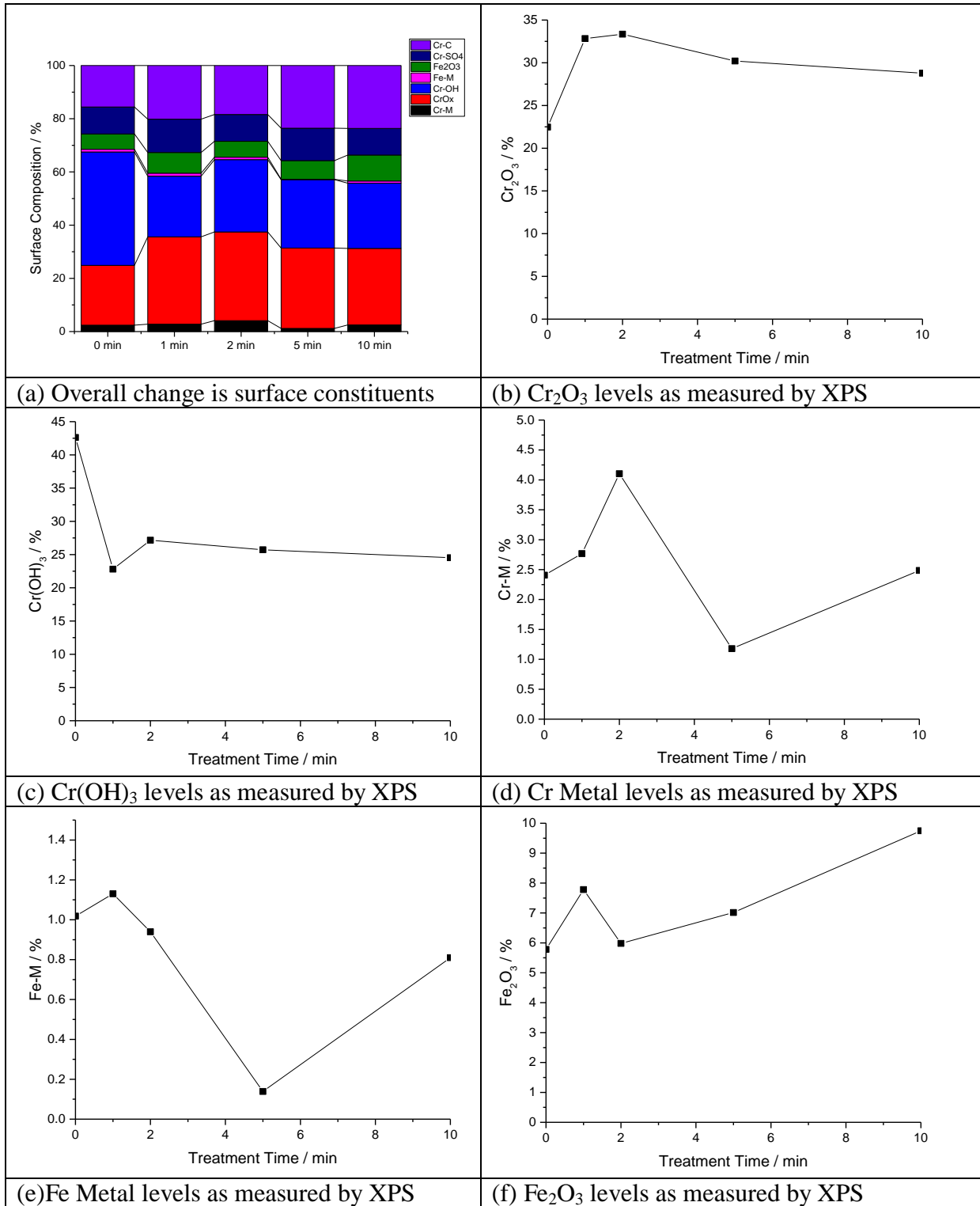

(a) Overall change is surface constituents	(b) Cr_2O_3 levels as measured by XPS
(c) $Cr(OH)_3$ levels as measured by XPS	(d) Cr Metal levels as measured by XPS
(e)Fe Metal levels as measured by XPS	(f) Fe_2O_3 levels as measured by XPS

Figure 3: Surface species as measured by XPS for 5% citric acid, 95 °C at each treatment time.

3.4 Surface Topology

Atomic force microscopy has been employed to assess the topological effects of the citric acid pre-treatment at the micron scale. Figure 4 gives a visual representation of the changing surface which arises as a result of the exposure to citric acid. Without treatment, the surface can be characterised as being smooth with low frequency bumps, Figure 4(a). There appears to minor craters on the surface which are attributed to pinholes in the chromium layer. Increasing the exposure to citric acid results in an increase in the surface roughness (R_a) and with 1 minute exposure resulting in a rougher with a similar underlying form with further exposure yielding a surface which is characterized by crevices and more abrupt surface features, Figure 4(e). It is not believed that the increase in surface roughness is a primary factor in improved adhesion seen in Figure 1, since further roughening leads to a reduction in adhesion and micro roughening has been shown to be insignificant in similar packaging materials, [23].
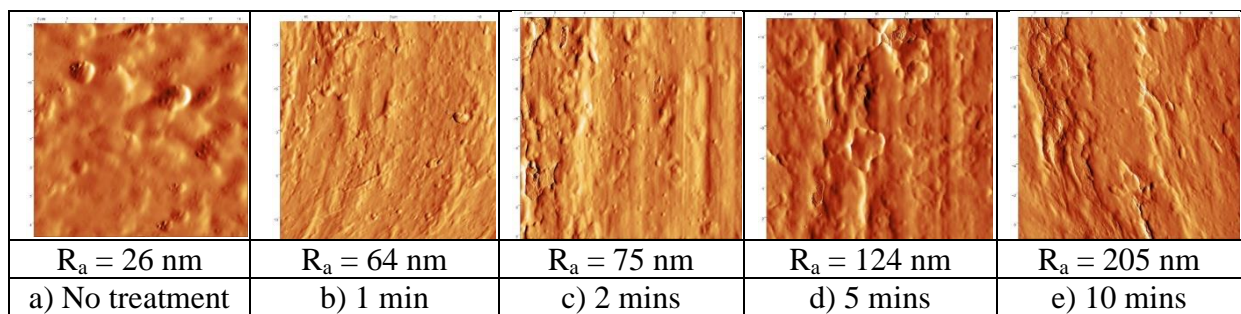
R_a = 26 nm	R_a = 64 nm	R_a = 75 nm	R_a = 124 nm	R_a = 205 nm
a) No treatment	b) 1 min	c) 2 mins	d) 5 mins	e) 10 mins

Figure 4: AFM images of the surface with changing treatment time.

3.6 Electrochemical characterization

The change in surface electrochemical activity is reinforced by the SVET measurements. When the untreated substrate is subjected to a 0.171 mol dm^{-3} NaCl solution, a number of discrete anodes and cathodes are seen at the substrate surface after 1 hour, Figure 5(a). As time progresses, the strength and number increases (Figure 5 b & c) until at 24 hours a few discrete anodic and cathodic areas are observed on the substrate, Figure 6d. A similar pattern in observed after the surface has been treated for a minute with citric acid, Figure 5 (e-h). After 5 minutes, there is very little anodic or cathodic activity on the surface to begin with, but as time passes a with a gradual single large area anode is produced on the surface, Figure 5 (i-l), although the magnitude of the current density remains low. At 10 minutes, larger areas of iron are exposed and this results in greater areas of cathodic and anodic activity on the surface, Figure 5(m-p). These larger area of exposed sub coating iron contrast to the random pinholes of iron which are observed with the untreated surface. This provides clear evidence that the citric acid has resulted in surface passivation, but that excessive exposure can result in sub coating iron being exposed.
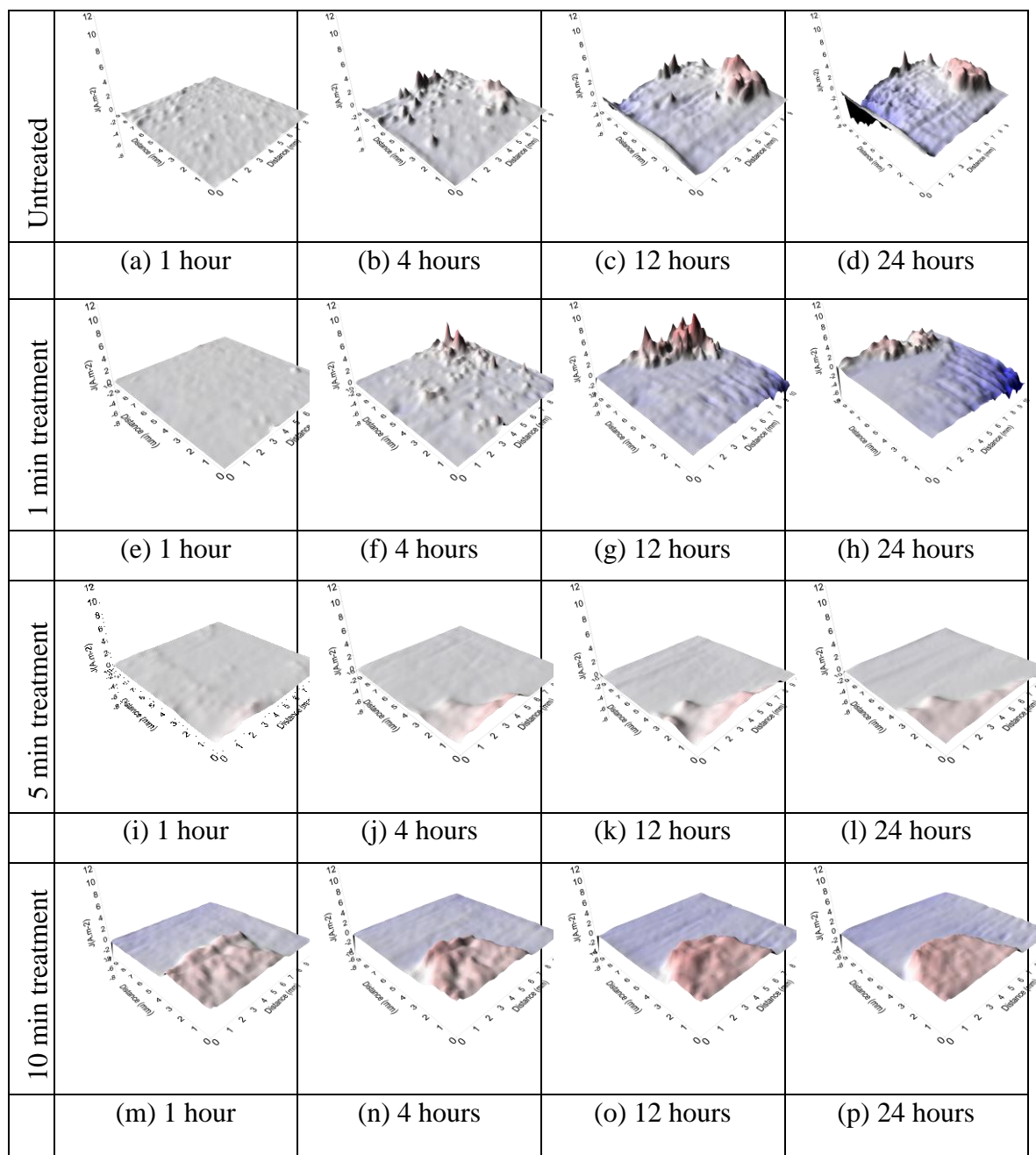
Figure 5 : SVET surface current map of each surface treatment time (a-d) untreated, (e-h) 1 minute citric pre- treatment, (i-l) 5 minute pre-treatment and (m-p) 10 minute pre-treatment at defined intervals (1, 4, 12 and 24 hours) during exposure to 0.171 mol dm^{-3} NaCl $_{(aq)}$.

Figure 6 shows a summary of the area-average, integrated SVET-derived anodic current density (J_{at}) versus time for the untreated and treated samples over a 24 hour period. The highest J_{at} values can be observed for the 10 minute pre-treatment sample, where from initial immersion at the start of the experiment, the anodic current is above 0.107 Am^{-2}. Untreated, 1 minute pre-treatment and 5 minute pre-treatment all start at low values for J_{at} , indicating a corrosively passive surface in the initial 0-2 hours. However as the immersion time is increased in the electrolyte the corrosion activity of untreated and 1 minute pre-treatment

rises to peaks of 0.202 Am^{-2} and 0.237 Am^{-2} respectively. At 5 minutes the anodic current values over a 24 hour period remain low, representing very little corrosion activity and a passive surface coating.
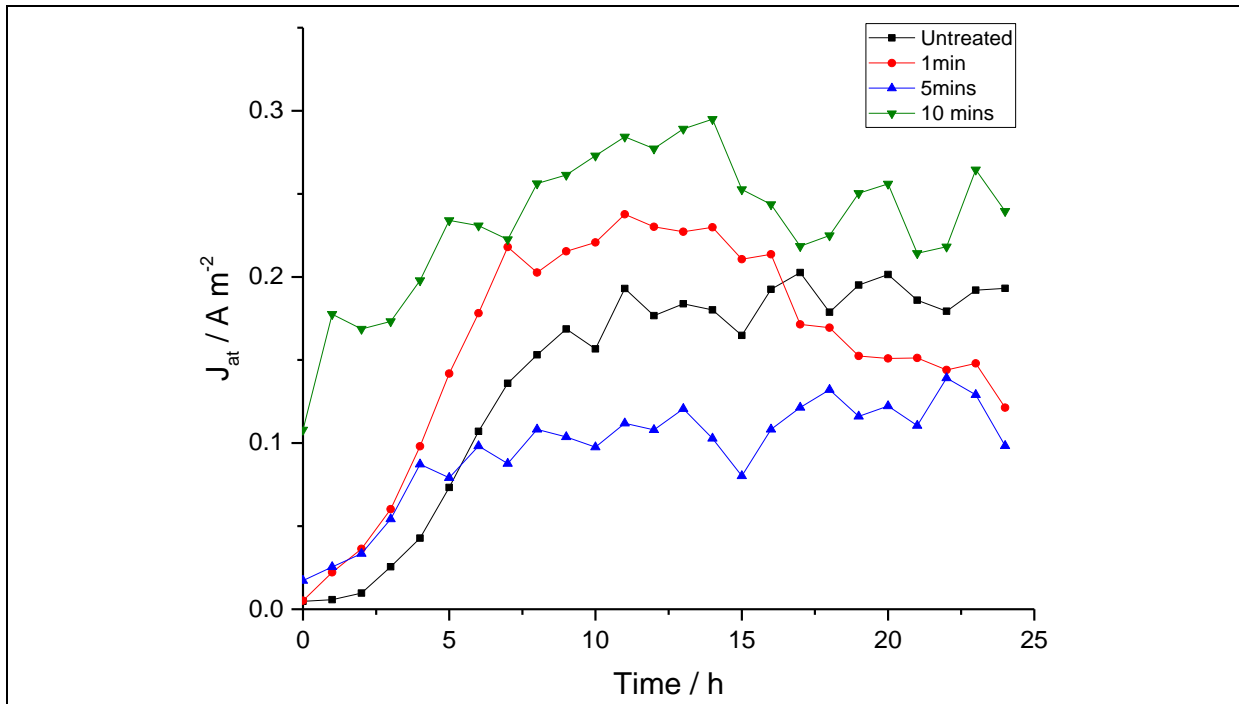

Figure 6 : Summary of area-averaged, integrated SVET-derived anodic current density versus time profiles obtained for untreated, 1 minute citric pre-treatment, 5 minute pre-treatment and 10 minute pre-treatment, immersed in aerated 0.171 mol dm^{-3} NaCl (aq)

4. Discussion

Given the processing times required in practice are typically in the order to seconds in order to maintain production speeds of 100m/min (1.7 m/s) or more then the application of this technique to Cr(III) coated packaging steel at the point of manufacture may be impractical. A 3m tank would result in an exposure time of less than 2 seconds which is insufficient to produce any positive improvement in performance. There would be little room for increasing the reaction rate in order to reduce exposure times as a further temperature rise beyond 95 °C would be impractical and the impact of increased concentration is limited. There is however an opportunity for its use for specialist downstream applications where processing speeds are lower.

The primary mechanism by which the citric acid improves adhesion on the surface is by the passive oxidation of the chromium metal to chromium oxide which provides a greater bonding sites for the lacquer. The window for optimum treatment of chromium coated packaging steel differs significantly from that used for pure stainless steel materials as there is a significant risk of over treatment. Any surface iron acts as a potential point of failure, exposing the subsurface iron in the substrate. The presence of any iron on the untreated substrate plays a key role in determining the efficacy and window of operation of surface deterioration during the sterilisation process. Any reduction in the surface iron will allow a wider operational window, allowing chromium metal to chromium oxide oxidation while limiting exposure of the subsurface. It is postulated that a reduction in surface iron will also reduce the time required for treatment as the chromium oxidation will become the only /

dominant oxidative mechanism. This mechanism up to 5 min treatment time appears to be the oxidation of chromium metal and chromium hydroxide to chromium oxide, as well as the dissolution of the iron species, reducing anodic activity on the surface of the substrate. In addition to this, the increase in surface roughness through the pre-treatment will provide a greater opportunity for a mechanical interlocking effect of the lacquer to the surface, as well as a greater surface area of adhesion promoting species for the lacquer to bond to.

5. Conclusion

The treatment of next generation packaging steels based on Cr(III) deposition chemistry with citric acid has been shown to improve the adhesion of lacquers during the subsequent sterilisation process. Through surface oxidation the adhesion of the lacquers is enhanced, but there exists an optimum window of citric acid treatment is dependent on both the acid concentration and temperature. Under optimum conditions, adhesion can be significantly improved but excessive treatment can lead to degradation in adhesion to pre-treatment levels. Improved homogeneity of the surface chromium coating are important in widening the effectiveness of the treatment and the overall system performance.

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