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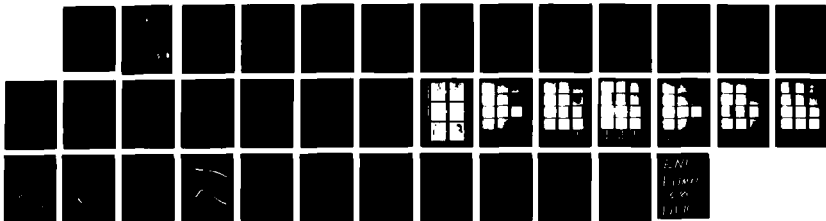
PRIMERLESS FINISHING SYSTEMS FOR THE CORROSION  
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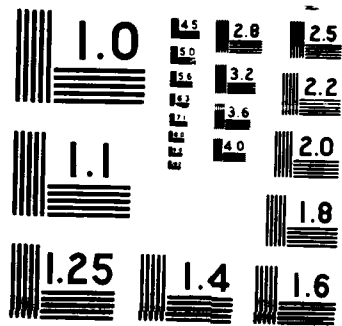
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**PRIMERLESS FINISHING SYSTEMS FOR THE  
CORROSION PROTECTION OF ALUMINUM**

Donald J. Hirst, Charles R. Hegedus, Stephen J. Spadafora,  
and Anthony T. Eng

Air Vehicle and Crew Systems Technology Department  
NAVAL AIR DEVELOPMENT CENTER  
Warminster, PA 18974-5000

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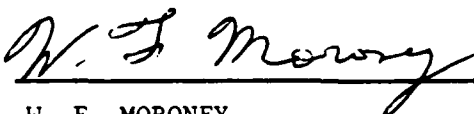
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19 ABSTRACT (Continue on reverse if necessary and identify by block number)  A trend to develop primerless systems for military equipment has begun in which a topcoat can be applied directly to surface treated aluminum. Two approaches to obtain a primerless system are currently available and were investigated. In the first approach, a standard sulfuric acid anodized surface was sealed with a polyurethane colloidal suspension. The resulting pretreatment can be directly topcoated. The second approach involved the application of a primerless or self-priming topcoat applied directly to a surface pretreatment. Adhesion, electrochemical impedance spectroscopy, flexibility and corrosion resistance tests were performed on several aluminum substrates to determine the physical properties of the coating-pretreatment systems. The results of this analysis indicate that these primerless systems show promise and should be considered for specific finishing system applications. <i>Keywords:</i>					
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## INTRODUCTION

The use of aluminum in the construction of military equipment is widespread due to its high specific strength compared to other structural alloys. This is vividly illustrated in airframe and aerospace structures where aluminum is by far the most commonly used material. Although structural and operational requirements are the primary concerns during design and construction of military equipment, component reliability and maximum lifetime with minimum maintenance are also required. A major influence on component performance is material properties. Materials, processing methods and protective pretreatments which minimize service failures must be utilized. Nowhere is this more apparent than with Navy aircraft which are usually stationed in highly corrosive environments.

In order to minimize the threat of deterioration, aluminum alloys are selected which have the required mechanical properties and exhibit less susceptibility to corrosive attack. Nonetheless, these alloys, if left unprotected, would rapidly corrode and cause the aircraft to be grounded. Therefore, inorganic surface treatments and organic coatings are specified for virtually all military equipment and especially aerospace systems. MIL-S-5002C, "Surface Treatments and Inorganic Coatings for Metal Surfaces of Weapons Systems" describes cleaning requirements and surface treatments for aluminum alloys. MIL-F-7179, "Finish, Coatings and Sealants for the Protection of Aerospace Weapons Systems" provides the requirements for paint and organic coatings used on U.S. military aircraft. References (1, 2) provide more detailed descriptions of corrosion control documents and finishing systems for military equipment.

In general, Navy aircraft finishing systems for aluminum consist of an inorganic surface treatment followed by a series of organic coatings. The surface treatment can produce either an anodized film or chromate conversion coating. The former is the product of an anodization and seal process which is performed in accordance with MIL-A-8625. The chromate conversion coating is achieved by applying materials conforming to MIL-C-81706 to produce a conversion coating meeting MIL-C-5541. The organic coating system consists of an epoxy primer (MIL-P-23377 or MIL-P-85582) and a polyurethane topcoat (MIL-C-83286 or MIL-C-85285). Several types of aircraft also require a coat of spray sealant (MIL-S-8802, MIL-S-81733, or MIL-P-87112) between the primer and topcoat. This finishing system was specifically designed to protect aluminum aircraft structures from the harsh aircraft carrier environment. The surface treatments enhance corrosion inhibition and adhesion of the subsequent coatings. The primers are adherent, and they inhibit corrosion of the substrata due to a high concentration of strontium chromate (3). The polyurethane topcoats are chemical and weather resistant, flexible and provide the required optical properties. A sealant coat is occasionally applied to enhance the flexibility of the coating system and prevent cracking of the paint, especially around fasteners and areas of excessive flexing.

Although the finishing system described above has been the premier finishing system on aircraft for 20 years, it has several deficiencies. The primer is brittle, especially at low operating temperatures (-51°C), resulting in cracking of the paint system on highly flexed areas. Sealants are soft and easily deformed and are difficult to apply and remove. In addition increased awareness and concern for environmental preservation and worker safety have caused local and state governments to limit volatile organic compound (VOC) emissions during painting operations. These regulations have impacted equipment manufacturers and rework depots by limiting the amount and types of paints which can be applied. The carcinogenic effects of chromates, which are used in conversion coatings and primers, present another concern about the current finishing system.

The issues listed above have prompted a recent trend to develop finishing systems which essentially consist of a surface pretreatment and one organic coating (4-7). This has been accomplished by using either a pretreatment, which can be directly coated with conventional topcoats (4), or a topcoat which can be applied to conventional inorganic pretreatments (5-6). In either case, the application of a primer is

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eliminated saving application time, manhours, and materials. The objective of this effort was to investigate the effectiveness of these systems to protect aluminum substrates.

### DESCRIPTION OF PRIMERLESS FINISHING SYSTEMS

As stated above, there are two approaches to eliminate the primer from a finishing system: (1) modify the inorganic pretreatment or (2) modify the topcoat. Reference (4) discloses a coating composition and application process for a modified anodized surface treatment which precludes the use of a subsequent primer prior to topcoating. The process follows the standard anodizing procedure (8) except for the final sealing step. Anodizing is a process by which the thickness of the natural oxide surface film, normally 1 to 5 nm, is increased to 0.5 to 100  $\mu\text{m}$ . This is accomplished by creating a cell in which the aluminum is anodic to another metal in an aqueous acid solution, commonly sulfuric or chromic acid. When current is passed through the cell, aluminum oxide is formed on the surface. As the process continues, oxide and hydroxide ions in the electrolyte solution diffuse and penetrate into the surface until they reach the aluminum-oxide interface. At this point they combine with aluminum ions, thus increasing the oxide layer thickness. This process is continued until equilibrium is reached which is dependent on the specific process variables. The anodized film consists of a non-porous underlying layer with a porous oxide structure on the surface. In order to increase the corrosion resistance of the film, the porous layer is closed by sealing with steam, hot water, or hot water solutions.

In contrast to the conventional process (4), the modified procedure utilizes a colloidal suspension of polyurethane resin to seal the porous oxide surface. Typically this suspension contains 7% solids in an alkaline solvent water bath. Upon contact with the aluminum surface, normally at 180°C, the solution induces film hydration and also impregnates the porous structure. The particle size of the colloidal suspension is designed to fit within the anodized surface structure. Upon completion of the sealing step, the specimen is exposed to air which allows curing and crosslinking of the polyurethane seal. The resulting film is water and solvent resistant, hard, flexible and corrosion resistant. A standard topcoat, MIL-C-83286 or equivalent, can be applied to this substrata one hour after removal from the sealing tank. Adhesion of the polyurethane topcoat is expected to be good due to the obvious chemical compatibility between pretreatment and topcoat. Specific formulations and procedures are provided in reference (4).

An alternative method for eliminating the need for a primer is to use a topcoat which is self-priming. Reference (5) describes the development and properties of one such coating. This coating can be applied directly to an aluminum substrata and provide the properties of the conventional primer and topcoat system designed for use on military aircraft. This coating can be applied to deoxidized, anodized or chromate conversion coated aluminum surfaces. It consists of a two component, aliphatic polyurethane binder with titanium dioxide, zinc molybdate, zinc phosphate, an organo-zinc salt, vesiculated polymer bead pigments. The polyurethane binder provides adhesion, flexibility, chemical and weather resistance. All of the pigments contribute to the film's opacity, however, the zinc molybdate, zinc phosphate, and organo-zinc salt are also corrosion inhibitors. This Self-Priming Topcoat exhibits good adhesion, corrosion inhibition, flexibility, chemical and weather resistance. The volatile organic compounds (VOC) content of the admixed material, which is suitable for airless spray, is 415 grams per liter of paint. If conventional air spray is desired for the application technique, this coating can be thinned with either 1,1,1 trichloroethane, which is currently exempt from emission regulations or standard urethane thinners.

### EXPERIMENTAL

The objective of this effort was to illustrate the effectiveness of primerless finishing systems for aluminum. In order to accomplish this, the two primerless systems described above and the standard

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paint system on Navy and Air Force aircraft (MIL-P-23377D and MIL-C-83286) were applied to bare and clad 2024 T-3 and 7075 T-6 aluminum alloys. All of the systems were evaluated for adhesion, chemical (fluid) resistance, flexibility and corrosion protection. The following is a description of the substrates, coatings, and experimental procedures utilized.

### Substrates and Coatings

Table 1 lists the twelve substrates and pretreatments which were utilized. The urethane sealed, sulfuric acid anodized (SAA) specimens were prepared by Lockheed Georgia, the dichromate sealed SAA specimens were prepared at our laboratories, and the chromate conversion coated specimens were obtained from Q Panel. The chromic acid and conversion coated specimens represent the common substrates found on military aircraft prior to painting. With the exception of the flexibility tests, all of the test procedures were conducted on all of these substrates. The flexibility tests were conducted on anodized 2024-O (annealed) aluminum specimens which were sealed with either the urethane colloidal suspension or hot water.

The three paint systems analyzed on all of the substrates in Table 1 are:

1. MIL-P-23377D, Type 1 "Primer Coatings, Epoxy Polyamide, Chemical and Solvent Resistant". Film thickness: 15.2 to 22.9 microns (0.006 to 0.009 inches). MIL-C-83286, "Coating Urethane, Aliphatic Isocyanate, for Aerospace Application". Film thickness: 50.8 to 55.9 microns (0.020 to 0.022 inches).
2. MIL-C-83286. Film thickness: 50.8 to 55.9 microns.
3. Self-Priming Topcoat (5). Film thickness: 50.8 to 55.9 microns.

The above coatings were applied by conventional air spray and were allowed to cure for seven days prior to testing.

### Experimental Procedures

#### Adhesion

Adhesion of the finishing systems was evaluated using two methods: wet tape adhesion (ASTM D 3359, method A) and scrape adhesion (ASTM D 2197, method A). The wet tape test was performed by immersing a specimen in distilled water for 24 hours. Upon removal, two parallel scribes, 1 inch apart, were cut through the coating and into the substrata. An "X" was subsequently scribed through the coating between the two initial scribes. A strip of 3M 250 masking tape was applied firmly to the coating surface perpendicular to the scribe lines and immediately removed with one quick motion. The specimens were examined for removal and uplifting of the coating from the substrata and the percentage of coating remaining on the surface was recorded.

The scrape test was performed on specimens with a section of the substrata surface exposed. The instrument used to perform this test was a SG-1605 Scrape Adhesion Test Apparatus manufactured by Gardner Laboratory. The test was performed by guiding a weighted stylus at a 45° angle to the specimen along the exposed substrata into the coating system. The scrape adhesion was recorded as the heaviest weight used without shearing the coating from the substrata.

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### Chemical (Fluid) Resistance

The ability of the finishing systems to resist common fluids used in aircraft was evaluated by immersing each system in lubricating oil, hydraulic fluid, a hydrocarbon solvent, and water under the conditions listed in Table 2. The coatings subsequently were examined for softening, upifting, blistering, and other defects which may have resulted from the exposure.

### Flexibility

The impact flexibility of the coating systems was evaluated at 23°C (74°F) using Method 6226 (G.E. Impact) of Federal Test Method Standard 141B. The test apparatus consisted of a solid steel cylinder weighing 1.69 kg (3.7 lbs) which has spherical knobs protruding from the end. These knobs are designed such that the coating system is subjected to elongations of 0.5, 1, 2, 5, 10, 20, 40, and 60%. The impact is accomplished by allowing the steel cylinder to fall freely from a height of 1.05 meters (42 inches) through a hollow cylinder guide, striking the reverse side of the specimen. The imprints formed from the knobs were examined and the impact elongation was recorded as the highest deformation without cracking of the coating.

The coating systems were also tested for flexibility at -51°C which is common for military aircraft cruising at high altitudes. The test method is described in ASTM D 1737 and is performed by bending the specimen 180° around 0.32, 0.63, 1.27, and 2.54 cm (1/8, 1/4, 1/2, and 1 inch) diameter mandrels. After returning to room temperature, the coating were examined for cracking along the bend. The most severe bend (smallest mandrel diameter) which the coating withstood without cracking was recorded.

### Corrosion Resistance

Four aluminum specimens of each finishing system were scribed in a figure "X" through the coating into the substrata. Two specimens each were exposed in 5% salt spray (ASTM B 117) for 2000 hours and two were exposed to SO<sub>2</sub>/salt spray (ASTM G 85) for 500 hours. The panels were then inspected for corrosion in the scribe area and blistering of the coating. Subsequently, one panel was chemically treated to remove the organic coating without disturbing the substrata and the specimen was examined for corrosion.

### Electrochemical Impedance Spectroscopy (EIS)

EIS measurements were made using an EG&G Princeton Applied Research Corp. (PARC) Model M368-4 AC Impedance System with Model 5208EC Lock-in Analyzer. The test cell used for this investigation consisted of a glass o-ring joint clamped onto a coated metal specimen as described in reference (9). The electrolyte used for specimen exposure was a 3.5% NaCl solution with a pH of 6. A total of nine coating and pretreatment systems on the 7075-T6 aluminum alloy substrata were selected for evaluation with EIS. These systems were based on combinations of the three surface pretreatments (SAA-urethane, SAA-dichromate, and chromate conversion coating) each with the three coating systems (epoxy-urethane, urethane, and the Self-Priming Topcoat). The specimens were exposed to the electrolyte solution for 1200 hours at room temperature and periodic impedance measurements were made over the test exposure time. The first series of tests were performed after 24 hours of exposure in order to allow the electrochemical system to reach equilibrium.

## RESULTS AND DISCUSSION

A primer is normally used in a coating system to prepare the surface to be painted for the application of a topcoat. In most cases this means the primer enhances the adhesion of the topcoat. In

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addition, because the primer is adjacent to the substrata, it is the primary corrosion inhibitor for the substrata. Therefore, the suspected weakest point in a primerless system would be the substrata-topcoat interface. The finishing systems were analyzed with special emphasis placed on surface interaction phenomena at this interface, primarily adhesion and corrosion. Figure 1 is a series of scanning electron micrographs taken at 10,000X of the three different pretreatments on both bare and clad 2024 aluminum. These photographs illustrate the micro-topography of the pretreatments, which will ultimately effect the interfacial properties between the organic and inorganic coatings.

The results of the adhesion tests are provided in Table 3. All of the scrape adhesion results are significantly higher than the standard 3 kg requirement for this property, indicating adequate adhesion under ambient laboratory conditions. In contrast, numerous systems failed the wet tape adhesion, indicating a susceptibility to coating-substrata disbondment upon exposure to water. Several conclusions can be drawn from these data. Systems with a conversion coating treatment performed well and this is expected since one objective of conversion coatings is to enhance adhesion of subsequent organic coatings. Many of the finishing systems containing both urethane and dichromate sealed anodized specimens had poor wet adhesion. This is not unusual. The sealing process improves corrosion protection because it minimizes the porosity of the anodized surface. However, in doing so, it leaves the surface with a smoother topography (Figure 1), minimizing the potential for mechanical adhesion. Bonding of the organic coating is then mainly dependent upon chemical bonds which are susceptible if water penetrates the coating and reaches the interface (10). Generally, the Self-Priming Topcoat exhibited the best overall performance in the wet tape test.

The flexibility test results are presented in Table 4. Standard specification criteria for these tests on low gloss coatings are 20% elongation and a 2 inch mandrel bend. All of the coating systems performed better on the water sealed anodized substrates than on the urethane sealed SAA. Since the same organic coatings were evaluated on both substrates, this indicates a deficiency at the SAA-urethane seal/coating interface. In addition, the urethane topcoat had slightly better impact elongation than the other two coating systems on both pretreatments. Poor flexibility is expected with the system containing the epoxy primer which is more brittle than the urethane, especially at low temperatures. However, the Self-Priming Topcoat has a polymer system which should provide as much flexibility as the standard topcoat. This is illustrated in the results for the mandrel bend test performed at -51°C where the Self-Priming Topcoat is much more flexible than the other two coatings. Although the lowest mandrel used was 1/4 inch, previous results (5) indicate the SPTC can withstand a 1/8 inch bend at this temperature without cracking.

Most of the coating systems exhibited excellent resistance to lubricating oil, hydraulic fluid, hydrocarbons and water. The Self-Priming Topcoat peeled from the urethane sealed SAA pretreatment after immersion in lubricating oil. This was unexpected considering the Self-Priming Topcoat has resisted these exposures on numerous substrates and that it has a similar polyurethane binder to MIL-C-83286 which showed no signs of failure. Another deficiency was observed with the MIL-C-83286 polyurethane topcoat on the urethane sealed, anodized panels when immersed in water for 4 days at 49°C. The coating had tiny blisters over the entire surface of the panel. Since no other system with the urethane pretreatment failed this test, this indicates a slight adhesion weakness at the coating-pretreatment interface. This weak adhesion, however, could improve with aging of the finishing system.

The specimens which were exposed to 5% salt spray for 2000 hours were examined for corrosion in the scribe area and for blistering of the coating. Subsequently the coatings were carefully removed from the surface using a chemical stripper without disturbing the underlying substrata. A summary of the evaluation is provided in Table 5 and photographs of the specimens with the coatings removed are provided in Figures 2-4. The condition of the specimens illustrated in these figures is indicative of the coating system performance. The standard epoxy primer-polyurethane topcoat performed well on all

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substrates. There were no significant corrosion products in the scribe or blistering of the coating. One specimen with the standard system on chromate conversion coated 7075 clad exhibited several pits along the scribe. In addition, slight uplifting of the standard coating system at the scribe was noticed on the urethane seal anodized specimens, however this was considered insignificant. The Self-Priming Topcoat also performed well on all of the substrates. There was no uplifting or blistering of the coating on any section of the specimens. The scribe areas had slight to moderate deposits of aluminum oxide with no pitting, however examination of these specimens after removing the coating indicated these products were minimal and confined to the scribe area. A previous report (5) indicated that these deposits are formed early during salt spray exposure but no further corrosion occurs for up to one year. This suggests that these deposits assist in the corrosion inhibition process.

The MIL-C-83286 polyurethane topcoat performed well on the dichromate sealed SAA with no blistering or uplifting of the coating and slight corrosion in the scribe area of the 7075 specimens. (Corrosion on the corner of the 2024 T-3 specimen in Figure 3 was the result of an edge effect and was discounted.) The good performance of the urethane on this substrata was unexpected because, aside from the dichromate seal which is damaged in the scribe area, there are no other corrosion inhibitors in the system. The polyurethane topcoat showed some corrosion products and pitting in the scribe of all four substrates treated with the sulfuric acid anodized-urethane seal. Performance on the chromate conversion coated pretreatment was poor. All four substrates showed pitting and corrosion along the scribe and corrosion of the substrata under the coating. We consider superficial corrosion products in the scribe to be acceptable, however any pitting in the scribe, corrosion extending from the scribe, or damage to the coating is unacceptable.

The specimens exposed to SO<sub>2</sub>/salt spray for 500 hours were also examined for damage to the coating and corrosion in and away from the scribe and these results are summarized in Table 6. Figures 5-7 are photographs of exposed specimens with the coatings removed. The SO<sub>2</sub>/salt spray environment simulates industrial stack gases such as those found on diesel powered carriers, and it is an extremely aggressive environment. The 500 hour exposure period was selected because differences in finishing system performance were observed after this duration. The specimens coated with the standard primer and topcoat system as well as those with the urethane topcoat had severe surface corrosion and/or pitting on all twelve substrates. In addition, the topcoat blistered on the conversion coating pretreatment and on the clad specimens with the SAA-dichromate seal. The extent of the corrosion with the topcoat was expected because of the lack of a corrosion inhibiting pigment. The results with the standard system were slightly unexpected since this system is considered one of the premier protective systems for aluminum due to the strontium chromate contained within the primer. The Self-Priming Topcoat outperformed the other two coating systems on all of the substrates. Although there were some slight spots of surface corrosion and small pits, these areas were barely noticeable and considered minor, relative to the extensive corrosion observed on the other specimens. Figures 5-7 provide vivid illustrations of the performance of all of the systems after sulfur dioxide/salt spray exposure.

Electrochemical impedance spectroscopy (EIS) provides qualitative and quantitative information about the corrosion resistance properties of both the coating and the substrata in addition to providing insight on the nature of their interfacial adhesion. Reference (11) provides a detailed description of EIS and its application for analyzing organic coating/metal substrata systems. Figures 8-11 contain Bode magnitude and phase diagrams of the EIS test results obtained at various exposure intervals for several of the coating/pretreatment systems. These specific spectra represent the significant EIS trends that were identified during this investigation.

After 24 hours immersion, the Self-Priming Topcoat (SPTC) on all three substrates had an impedance of  $1.2 \times 10^9$  in the low frequency range ( $10^{-2}$  Hz) which was the highest impedance of the three coatings as displayed in Figure 8. High impedance values correlate to coatings with low

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conductivity that provide good barrier protection to the substrates to which they are applied. This impedance value is far above  $10^7$  ohms which is widely accepted as the lower limit below which no barrier protection is provided by the coating (11). In the high frequency range, the SPTC had phase angles between  $-80^\circ$  and  $-90^\circ$ . These phase angles also indicate a good barrier coating, where  $-90^\circ$  would be a perfect capacitor/barrier. In addition, the shape of the curve for the impedance of the SPTC is virtually straight over most of the frequency range with a negative slope, again indicating capacitive behavior (i.e. good barrier properties). The same results were observed for the SPTC throughout the 1200 hour test duration as illustrated in Figure 9. For all pretreatments, the low frequency SPTC impedance remained above  $10^9$  ohms, while the high frequency phase angles continued to exhibit capacitive behavior, remaining between  $-75^\circ$  and  $-90^\circ$ . There was a small shift in the low frequency phase angle curve for the SAA-urethane/SPTC system over the test duration. The change from capacitive to resistive behavior had shifted slightly to the left (lower frequency) indicating better barrier properties. This change could have resulted from several sources. One possible explanation is decreased micropore size within the coating, caused by swelling of the polymer. However, since the phenomenon occurred with only one pretreatment, this explanation was dismissed. Another plausible explanation is increased coating adhesion. This increased degree of interfacial bonding with time, also noted in reference (12), could have been enhanced or catalyzed by the presence of some electrolyte at the interface. Finally, during the 1200 hour test period, the chemical corrosion resistance properties of the inhibitors in this coating did not come into play and will not be addressed here.

The primer and topcoat (EP-UR) system on all three substrates had an impedance of  $6.3 \times 10^7$  ohms after 24 hours immersion. Again this indicates barrier type properties, however not as good as the SPTC. This lower barrier protection is also apparent in the phase diagram where the phase angles for the EP-UR system were between  $-60^\circ$  and  $-80^\circ$ , showing less capacitive behavior. The shape of the EP-UR magnitude curve was similar to the SPTC curve, however it was evident at a lower impedance range. The improved barrier protection provided by the Self-Priming Topcoat resulted because this coating was specifically designed to have high flexibility and a smooth surface and therefore it is less porous than the standard epoxy-urethane coating system. In addition, the EP-UR impedance curve leveled off at a higher frequency than the SPTC, indicating lesser barrier properties. Although the EP-UR coating is not as good a barrier coating as the SPTC, it does provide excellent corrosion protection as indicated in the salt spray results and reference (3). The low frequency impedance magnitude of the EP-UR system remained between  $10^7$  and  $10^8$  ohms and the shape of the curve was similar over most of the exposure time. However at 1200 hours, the chromate conversion coating/EP-UR impedance curve began to show an upward turn after leveling off in the low frequency range as shown in Figure 10. Also, the phase angle curve was beginning to develop a peak in the high frequency range. These two trends indicate the presence of electrolyte at the interface resulting from some adhesion loss. Also, some type of electrochemical reactions were occurring at the interface, probably corresponding to chemical inhibition of the corrosion process by the inhibitors within the primer.

The unprimed polyurethane topcoat (UR) performed differently with the various pretreatments. After one day, the UR with the SAA/dichromate seal and the conversion coated pretreatments had low impedance values ( $1.3 \times 10^7$  and  $2.5 \times 10^6$  ohms, respectively) and provided little or no barrier protection to the substrata. Furthermore, unlike the shape of the other two impedance curves, the UR impedance curve leveled off in the mid-frequency range and then began to curve upward at the low frequency range. This behavior corresponds to a porous coating with poor adhesion, where electrolyte is allowed to penetrate the film and accumulate at the coating-metal interface. Finally, the phase angle behavior for these pretreatments was significantly different than the other two materials (see Figure 11). At  $10^5$  Hz the phase angle was  $-80^\circ$ , however as the frequency decreased, the phase angle reached a maximum of approximately  $-5^\circ$  at about 1 Hz. This gradual change from capacitance to resistance then sharply reversed back to capacitance again in the low frequency area. This response indicates a double layer capacitance at the metal surface resulting from the presence of electrolyte at the interface caused by

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coating adhesion loss. As exposure continued, the inflections in the impedance curves for the UR with the SAA-dichromate seal and chromate conversion coating pretreatments began to shift to higher frequencies with impedance values below  $10^7$  showing no real barrier protection. Also, the peak maximum in the phase angle diagram for the conversion coated specimens shifted from 1 Hz at 24 hours to 10 Hz at 504 hours and finally reached 50 Hz at 1200 hours. In addition to shifting, the peak broadened from spanning five decades to six decades and finally spanning seven decades, respectively. These changes indicate that electrochemical reactions were occurring at the interface and possibly represented the onset and propagation of the corrosion process.

The UR and SAA-polyurethane seal system performed closer to the other two coatings in the impedance diagram with an initial low frequency impedance of  $3 \times 10^8$  ohms and a virtually straight magnitude curve. This curve did not significantly change over the test duration. The phase angle curve for this system behaved like the SPTC and standard EP-UR systems in the low frequency range. However, in the high frequency area there was a resistance peak similar to the one described for the conversion coating-EP-UR system at 1200 hours. This peak gradually became more resistive, which relates to interface degradation.

The SPTC was by far the best barrier coating as demonstrated by EIS. The standard coating system also offered barrier protection to the substrates but not quite as good as the SPTC. The chemical protection provided by the corrosion inhibitors in the SPTC coating did not come into play during this test time. Similarly, the chemical corrosion inhibition of the EP-UR coating was not specifically demonstrated in the EIS tests, except possibly in the 1200 hour conversion coating results. The UR system provided poor barrier protection, with the exception of the polyurethane sealed SAA pretreatment, and offered no chemical protection against corrosion. Finally, as exposure time increased the results for the EP-UR and UR coatings changed for the different pretreatments, while the SPTC spectra remained virtually the same for all pretreatments over the entire 1200 hour test duration.

### SUMMARY

Comparison of all the performance data for the organic coatings indicated some correlation between the test results. Adhesion data and water resistance show a general trend in adhesion performance from the Self-Priming Topcoat as the best system, to the unprimed polyurethane topcoat as the worst. Likewise, the  $\text{SO}_2$ -salt spray test data and the EIS data correlated well with the adhesion data, again resulting in the same performance trend for the coating systems. The only exception was in the 5% NaCl salt spray test results. Here, the standard primer/topcoat (EP-UR) system properties proved to be slightly better than those of the Self-Priming Topcoat, but not to a significant degree.

In summarizing the performance of the inorganic pretreatments, the SAA-urethane resulted in poor adhesion and flexibility for nearly all of the alloy/organic coatings analyzed. However, it did assist in providing fair corrosion protection in salt spray and  $\text{SO}_2$ -salt spray. Its electrochemical impedance characteristics were promising when coated with the standard urethane topcoat. The SAA-dichromate provided fair adhesion for the urethane coatings but poor adhesion for the epoxy-urethane system. Its performance with all of the organic coatings was good in salt spray and fair in  $\text{SO}_2$ -salt spray. The chromate conversion coating provided excellent adhesion for all of the coatings. It also provided good corrosion protection in the salt spray when coated with either the standard system or the Self-Priming Topcoat. Performance in the  $\text{SO}_2$ -salt spray was only good when coated with the SPTC. The EIS data indicated that the chromate conversion coating appeared to be the best pretreatment for short term durations, however, its long term durability was significantly inferior to the SAA treatments. There was some disagreement between the results for the Electrochemical Impedance Spectroscopy and the wet tape adhesion tests. These differences in performance were attributed to the decreased thermodynamic

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activity of water in the ionic solution used in the EIS testing as apposed to distilled water used in the tape test (10).

The objective of this paper was to investigate primerless finishing systems in lieu of the standard primer and topcoat system currently used on military aircraft. Two alternatives to obtain a primerless system were evaluated: (1) Modification of the surface treatment and (2) Modification of the organic coating. The sulfuric acid anodized-urethane seal did not perform as well as the standard system. However it did show promising results in the electrochemical impedance analysis. The concept of having a pretreatment which forms strong chemical bonds with an applied organic coating is viable. Incorporating corrosion inhibitors in the subsequent coating may be essential to improve the overall corrosion protection, especially if the coating system is damaged. The Self-Priming Topcoat performed well throughout the evaluation. This alternative can be used over a variety of substrates. As previous mentioned, numerous advantages can be realized with the use of primerless finishing systems. Some of these advantages depend on the specific system and its application parameters. However, three advantages which would be prevalent with all approaches are:

1. Reduced volatile organic emissions.
2. Reduced chromate emissions.
3. Reduced finishing system application time.

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**TABLE 1: SUBSTRATES AND PRETREATMENTS FOR PRIMERLESS COATING SYSTEMS**

<u>ALLOY</u>	<u>PRETREATMENT</u>		
	<u>SAA-URETHANE SEAL</u>	<u>SAA-DICHROMATE SEAL</u>	<u>CHROMATE CONV COAT</u>
2024 T-3, BARE	XXX	XXX	XXX
2024 T-3, CLAD	XXX	XXX	XXX
7075 T-6, BARE	XXX	XXX	XXX
7075 T-6, CLAD	XXX	XXX	XXX

**TABLE 2: EXPOSURE CONDITIONS FOR CHEMICAL RESISTANCE EVALUATION**

<u>FLUID</u>	<u>IMMERSION PERIOD</u> <u>(HOURS)</u>	<u>TEMPERATURE</u> <u>(°C)</u>
LUBRICATING OIL	24	65
ENGINE OIL	24	121
WATER	96	49
HYDROCARBON	24	23

TABLE 3: ADHESION TEST RESULTS

SUBSTRATE	PRETREATMENT	COATING	WET TAPE (% REMAINING)	SCRAPE (KG)
2024 BARE	SAA-URETHANE	EP-UR*	97	9
2024 CLAD	SAA-URETHANE	EP-UR	10	7
7075 BARE	SAA-URETHANE	EP-UR	25	8
7075 CLAD	SAA-URETHANE	EP-UR	100	10
2024 BARE	SAA-URETHANE	UR	0	7
2024 CLAD	SAA-URETHANE	UR	0	4
7075 BARE	SAA-URETHANE	UR	0	6
7075 CLAD	SAA-URETHANE	UR	90	9
2024 BARE	SAA-URETHANE	SPTC	100	10
2024 CLAD	SAA-URETHANE	SPTC	0	9
7075 BARE	SAA-URETHANE	SPTC	75	10
7075 CLAD	SAA-URETHANE	SPTC	100	9
2024 BARE	SAA-DICHROMATE	EP-UR	80	9
2024 CLAD	SAA-DICHROMATE	EP-UR	0	5
7075 BARE	SAA-DICHROMATE	EP-UR	75	8
7075 CLAD	SAA-DICHROMATE	EP-UR	0	7
2024 BARE	SAA-DICHROMATE	UR	100	10
2024 CLAD	SAA-DICHROMATE	UR	0	7
7075 BARE	SAA-DICHROMATE	UR	100	10
7075 CLAD	SAA-DICHROMATE	UR	100	10
2024 BARE	SAA-DICHROMATE	SPTC	100	10
2024 CLAD	SAA-DICHROMATE	SPTC	100	10
7075 BARE	SAA-DICHROMATE	SPTC	100	10
7075 CLAD	SAA-DICHROMATE	SPTC	100	9
2024 BARE	CHR CONV COAT	EP-UR	100	10
2024 CLAD	CHR CONV COAT	EP-UR	100	10
7075 BARE	CHR CONV COAT	EP-UR	100	10
7075 CLAD	CHR CONV COAT	EP-UR	100	10
2024 BARE	CHR CONV COAT	UR	100	10
2024 CLAD	CHR CONV COAT	UR	100	10
7075 BARE	CHR CONV COAT	UR	70	10
7075 CLAD	CHR CONV COAT	UR	100	10
2024 BARE	CHR CONV COAT	SPTC	100	10
2024 CLAD	CHR CONV COAT	SPTC	100	10
7075 BARE	CHR CONV COAT	SPTC	100	10
7075 CLAD	CHR CONV COAT	SPTC	100	10

\*EP-UR; MIL-P-23377 PRIMER AND MIL-C-83286 TOPCOAT  
 UR; MIL-C-83286 TOPCOAT  
 SPTC; SELF-PRIMING TOPCOAT

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TABLE 4: FLEXIBILITY TEST RESULTS

<u>PRETREATMENT/COATING</u>	<u>IMPACT @ 23°C (% ELONGATION)</u>	<u>MANDREL BEND @ -51°C (MANDREL DIAM., INCHES)</u>
SAA-WATER/EP-UR	20	1
SAA-WATER/UR	40	1
SAA-WATER/SPTC	20	1/4
SAA-URETHANE/EP-UR	20	>1
SAA-URETHANE/UR	40	>1
SAA-URETHANE/SPTC	5	1/2

TABLE 5: SUMMARY OF SALT SPRAY EXPOSURE RESULTS

<u>SUBSTRATE</u>	<u>PRETREATMENT</u>	<u>CORATING</u>	<u>COMMENTS</u>
2024 BARE	SAA-URETHANE	EP-UR*	NO CORROSION
2024 CLAD	SAA-URETHANE	EP-UR	NO CORROSION
7075 BARE	SAA-URETHANE	EP-UR	NO CORROSION
7075 CLAD	SAA-URETHANE	EP-UR	SLIGHT SURFACE CORROSION PRODUCTS IN ENTIRE SCRIBE
2024 BARE	SAA-URETHANE	UR	EXTENSIVE PITTING IN SCRIBE, CORROSION EXTENDING 1/8 INCH FROM SCRIBE
2024 CLAD	SAA-URETHANE	UR	SURFACE CORROSION PRODUCTS IN ENTIRE SCRIBE; SEVERAL PITS ALONG SCRIBE
7075 BARE	SAA-URETHANE	UR	EXTENSIVE PITTING IN SCRIBE
7075 CLAD	SAA-URETHANE	UR	SURFACE CORROSION PRODUCTS IN ENTIRE SCRIBE; SEVERAL PITS ALONG SCRIBE
2024 BARE	SAA-URETHANE	SPTC	SURFACE CORROSION PRODUCTS IN 50% OF SCRIBE
2024 CLAD	SAA-URETHANE	SPTC	SLIGHT SURFACE CORROSION PRODUCTS IN ENTIRE SCRIBE
7075 BARE	SAA-URETHANE	SPTC	SURFACE CORROSION PRODUCTS IN ENTIRE SCRIBE
7075 CLAD	SAA-URETHANE	SPTC	SURFACE CORROSION PRODUCTS IN ENTIRE SCRIBE
2024 BARE	SAA-DICHROMATE	EP-UR	NO CORROSION
2024 CLAD	SAA-DICHROMATE	EP-UR	NO CORROSION
7075 BARE	SAA-DICHROMATE	EP-UR	NO CORROSION
7075 CLAD	SAA-DICHROMATE	EP-UR	NO CORROSION
2024 BARE	SAA-DICHROMATE	UR	NO CORROSION
2024 CLAD	SAA-DICHROMATE	UR	NO CORROSION
7075 BARE	SAA-DICHROMATE	UR	SLIGHT SURFACE CORROSION PRODUCTS IN ENTIRE SCRIBE
7075 CLAD	SAA-DICHROMATE	UR	SLIGHT SURFACE CORROSION PRODUCTS IN ENTIRE SCRIBE
2024 BARE	SAA-DICHROMATE	SPTC	NO CORROSION
2024 CLAD	SAA-DICHROMATE	SPTC	SLIGHT SURFACE CORROSION PRODUCTS IN 50% OF SCRIBE
7075 BARE	SAA-DICHROMATE	SPTC	SLIGHT SURFACE CORROSION PRODUCTS IN 25% OF SCRIBE
7075 CLAD	SAA-DICHROMATE	SPTC	SLIGHT SURFACE CORROSION PRODUCTS IN 75% OF SCRIBE
2024 BARE	CHR CONV COAT	EP-UR	NO CORROSION
2024 CLAD	CHR CONV COAT	EP-UR	NO CORROSION
7075 BARE	CHR CONV COAT	EP-UR	NO CORROSION
7075 CLAD	CHR CONV COAT	EP-UR	SLIGHT SURFACE CORROSION IN ENTIRE SCRIBE; SEVERAL PITS ALONG SCRIBE
2024 BARE	CHR CONV COAT	UR	LARGE SEVERE PITS ALONG SCRIBE; SCATTERED PITTING OVER ENTIRE SURFACE
2024 CLAD	CHR CONV COAT	UR	SURFACE CORROSION PRODUCTS IN SCRIBE INCLUDING 2 SEVERE SPOTS
7075 BARE	CHR CONV COAT	UR	SURFACE CORROSION/PITTING ALONG SCRIBE; SCATTERED PITTING OVER SURFACE
7075 CLAD	CHR CONV COAT	UR	SURFACE CORROSION AND PITTING ALONG SCRIBE; A FEW PITS UNDER COATING
2024 BARE	CHR CONV COAT	SPTC	SLIGHT SURFACE CORROSION IN SCRIBE; 2 SMALL AREAS EXTENDING FROM SCRIBE
2024 CLAD	CHR CONV COAT	SPTC	SLIGHT SURFACE CORROSION IN SCRIBE, 1 SMALL AREA EXTENDING FROM SCRIBE
7075 BARE	CHR CONV COAT	SPTC	SLIGHT SURFACE CORROSION IN THE SCRIBE; 2 SMALL AREAS ALONG SCRIBE
7075 CLAD	CHR CONV COAT	SPTC	SLIGHT SURFACE CORROSION IN THE SCRIBE

\*EP-UR: MIL-P-23377 PRIMER AND MIL-C-83286 TOPCOAT

UR: MIL-C-83286 TOPCOAT

SPTC: SELF-PRIMING TOPCOAT

TABLE 6: SUMMARY OF SULFUR DIOXIDE-SALT SPRAY EXPOSURE RESULTS

<u>SUBSTRATE</u>	<u>PRETREATMENT</u>	<u>COATING</u>	<u>COMMENTS</u>
2024 BARE	SAA-URETHANE	EP-UR*	PITTING IN ENTIRE SCRIBE
2024 CLAD	SAA-URETHANE	EP-UR	PITTING IN ENTIRE SCRIBE; SEVERE CORROSION EXTENDING 1/2" FROM SCRIBE
7075 BARE	SAA-URETHANE	EP-UR	PITTING IN AND EXTENDING FROM THE SCRIBE
7075 CLAD	SAA-URETHANE	EP-UR	SEVERE CORROSION IN AND EXTENDING 1/4" FROM SCRIBE
2024 BARE	SAA-URETHANE	UR	PITTING IN ENTIRE SCRIBE
2024 CLAD	SAA-URETHANE	UR	PITTING IN ENTIRE SCRIBE; SEVERE CORROSION EXTENDING 1/2" FROM SCRIBE
7075 BARE	SAA-URETHANE	UR	PITTING IN AND EXTENDING FROM THE SCRIBE
7075 CLAD	SAA-URETHANE	UR	SEVERE CORROSION IN AND EXTENDING 1/4" FROM SCRIBE
2024 BARE	SAA-URETHANE	SPTC	NO CORROSION
2024 CLAD	SAA-URETHANE	SPTC	CORROSION ALONG 10% OF SCRIBE
7075 BARE	SAA-URETHANE	SPTC	NO CORROSION
7075 CLAD	SAA-URETHANE	SPTC	SEVERAL SLIGHT CORROSION SPOTS ALONG SCRIBE
2024 BARE	SAA-DICHROMATE	EP-UR	PITTING IN ENTIRE SCRIBE
2024 CLAD	SAA-DICHROMATE	EP-UR	PITTING IN AND EXTENDING 1/8" FROM ENTIRE SCRIBE
7075 BARE	SAA-DICHROMATE	EP-UR	PITTING IN AND EXTENDING FROM THE SCRIBE
7075 CLAD	SAA-DICHROMATE	EP-UR	SURFACE CORROSION AND PITTING IN AND EXTENDING 3/4" FROM SCRIBE
2024 BARE	SAA-DICHROMATE	UR	PITTING IN ENTIRE SCRIBE
2024 CLAD	SAA-DICHROMATE	UR	PITTING IN AND EXTENDING 1/8" FROM SCRIBE; PAINT BLISTERED
7075 BARE	SAA-DICHROMATE	UR	PITTING IN ENTIRE SCRIBE
7075 CLAD	SAA-DICHROMATE	UR	SURFACE CORROSION/PITTING EXTENDING 3/4" FROM SCRIBE; PAINT BLISTERED
2024 BARE	SAA-DICHROMATE	SPTC	NO CORROSION
2024 CLAD	SAA-DICHROMATE	SPTC	2 SMALL PITS ALONG SCRIBE
7075 BARE	SAA-DICHROMATE	SPTC	NO CORROSION
7075 CLAD	SAA-DICHROMATE	SPTC	SEVERAL SMALL PITS ALONG SCRIBE
2024 BARE	CHR CONV COAT	EP-UR	PITTING IN ENTIRE SCRIBE
2024 CLAD	CHR CONV COAT	EP-UR	PITTING IN AND EXTENDING SLIGHTLY FROM SCRIBE
7075 BARE	CHR CONV COAT	EP-UR	PITTING IN AND EXTENDING 1/4" FROM SCRIBE
7075 CLAD	CHR CONV COAT	EP-UR	SURFACE CORROSION AND PITTING IN AND EXTENDING 1/2" FROM SCRIBE
2024 BARE	CHR CONV COAT	UR	EXTENSIVE SURFACE CORROSION/PITTING OVER ENTIRE SURFACE; PAINT BLISTERED
2024 CLAD	CHR CONV COAT	UR	EXTENSIVE SURFACE CORROSION/PITTING OVER ENTIRE SURFACE; PAINT BLISTERED
7075 BARE	CHR CONV COAT	UR	EXTENSIVE SURFACE CORROSION/PITTING OVER ENTIRE SURFACE; PAINT BLISTERED
7075 CLAD	CHR CONV COAT	UR	EXTENSIVE SURFACE CORROSION/PITTING OVER ENTIRE SURFACE; PAINT BLISTERED
2024 BARE	CHR CONV COAT	SPTC	3 SMALL PITS ALONG SCRIBE
2024 CLAD	CHR CONV COAT	SPTC	SEVERAL SMALL PITS ALONG SCRIBE
7075 BARE	CHR CONV COAT	SPTC	5 SMALL PITS ALONG SCRIBE
7075 CLAD	CHR CONV COAT	SPTC	2 SMALL PITS ALONG SCRIBE

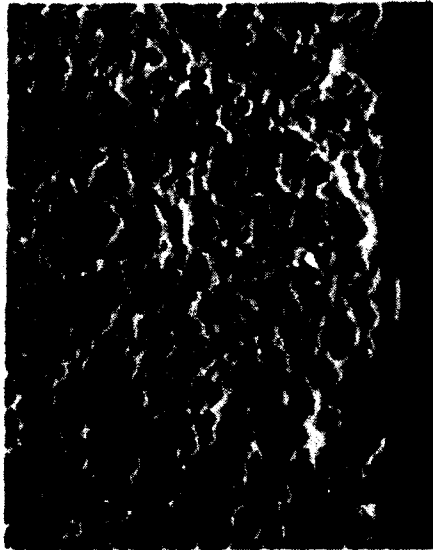
\*EP-UR: MIL-P-23377 PRIMER AND MIL-C-83286 TOPCOAT  
 UR: MIL-C-83286 TOPCOAT  
 SPTC: SELF-PRIMING TOPCOAT

**SAA-URETHANE SEAL**



**BARE**

**SAA-DICHROMATE SEAL**



**BARE**

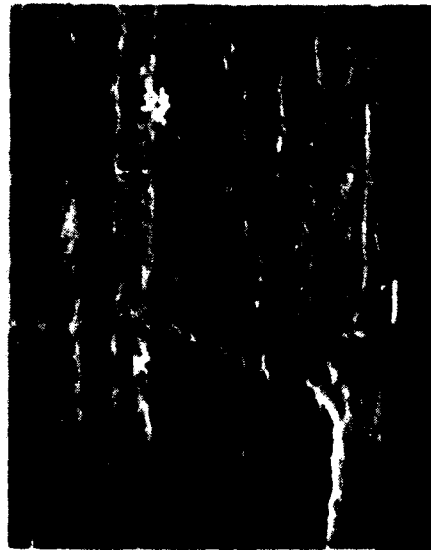
**CHROMATE CONV. COAT.**



**BARE**



**CLAD**



**CLAD**



**CLAD**

**FIGURE 1: SURFACE TOPOGRAPHY OF PRETREATMENTS (10000X).**

3024 T-0 BAKE

3024 T-0 CURE

NADC-88107-60

MIL-C-83286

MIL-P-23377

a

MIL-C-83286

N A D C  
SELF PRIMING  
TOPCOAT

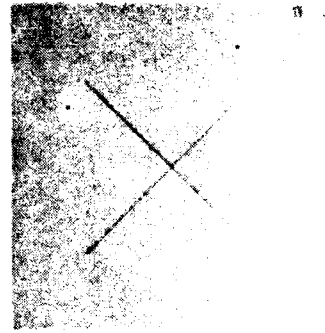
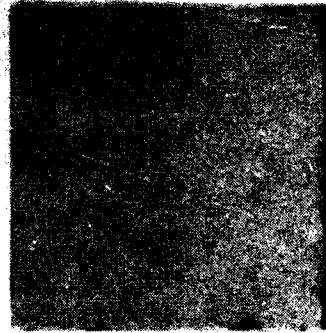
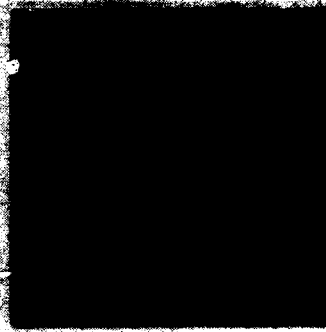
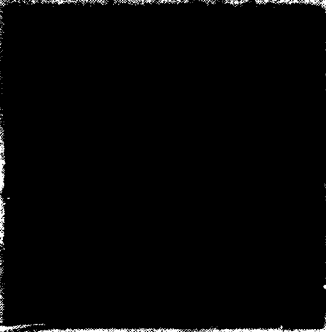
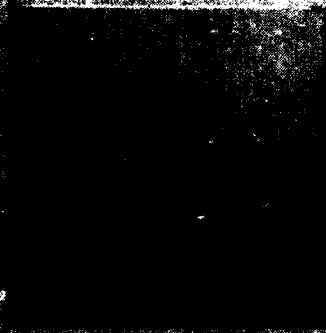
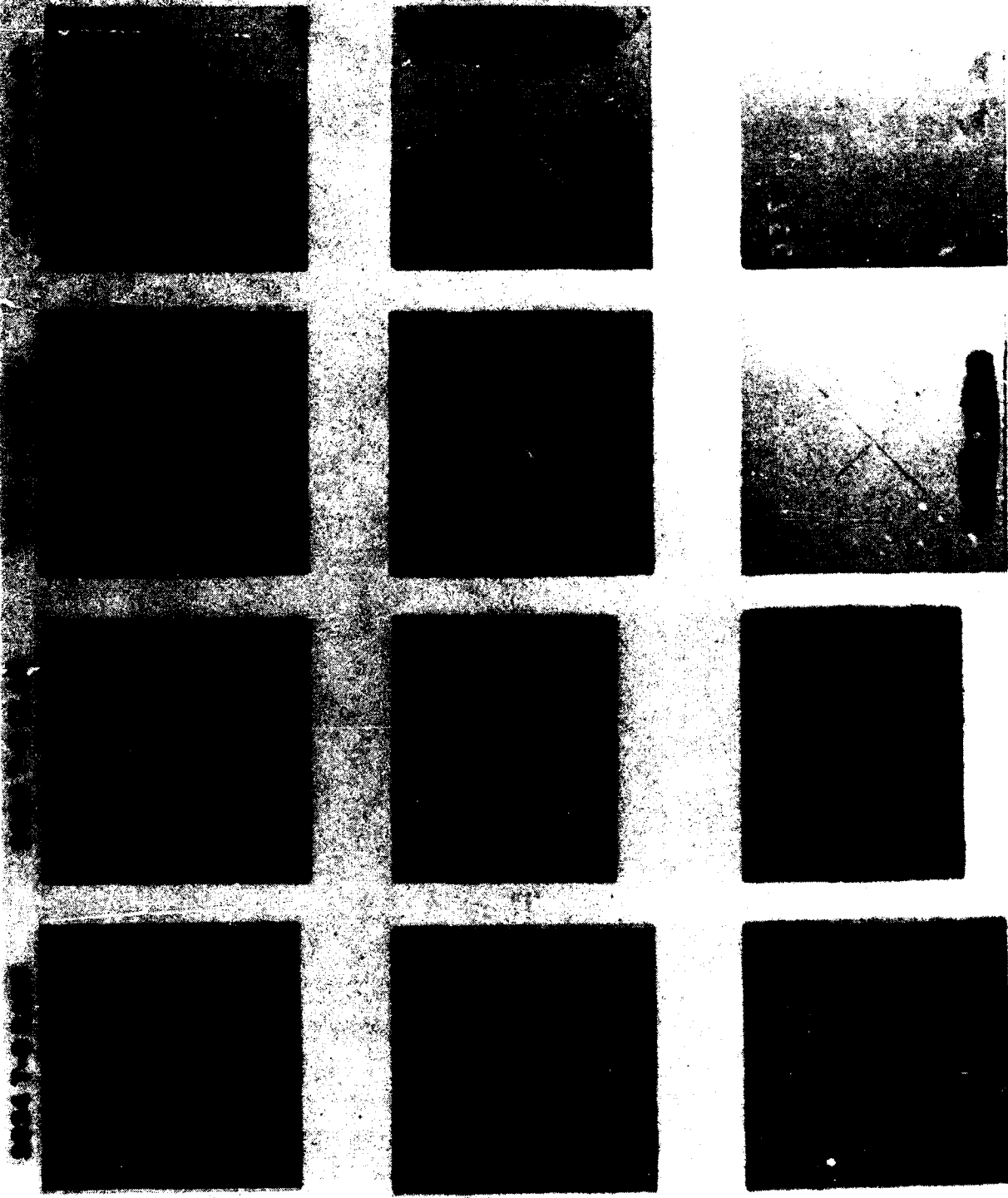


FIGURE 2: SULFURIC ACID ANODIZED-URETHANE SEALED SPECIMENS EXPOSED TO 5% SALT SPRAY FOR 2000 HOURS.

NADC-65107-60



ML-C-83286

ML-P-23377

&

ML-C-83286

N A D C  
SELF PRIMING  
TOPCOAT

FIGURE 3: SULFURIC ACID ANODIZED-DICHROMATE SEALED SPECIMENS EXPOSED TO 5% SALT SPRAY FOR 2000 HOURS.

NADC-88187-80

ML-C-83288

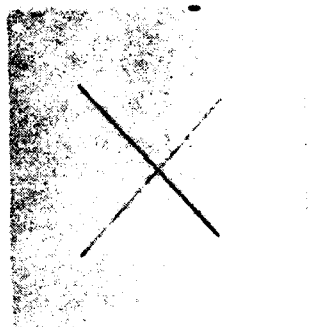
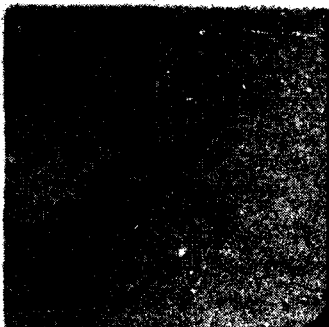
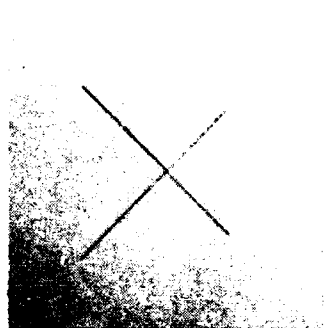
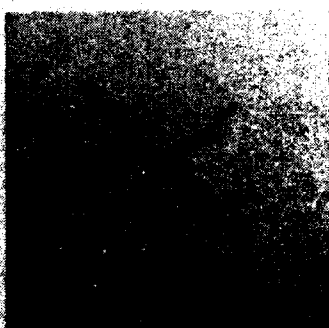
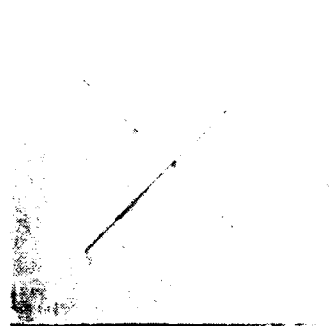
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ML-C-83288

N A D C  
SELF PRIMING  
TOPCOAT

FIGURE 4: CHROMATE CONVERSION COATED SPECIMENS EXPOSED TO 5% SALT SPRAY FOR 2000 HOURS.

NADC-88107-60



ML-C-83286

ML-P-23377

&

ML-C-83286

N A D C  
SELF PRIMING  
TOPCOAT

FIGURE 5: SULFURIC ACID ANODIZED-URETHANE SEALED SPECIMENS  
EXPOSED TO SULFUR DIOXIDE-SALT SPRAY FOR 500 HOURS.

2024 T-3 BASE



ML-C-83286



ML-P-23377  
&  
ML-C-83286



N A D C  
SELF PRIMING  
TOPCOAT

NADC-88107-60

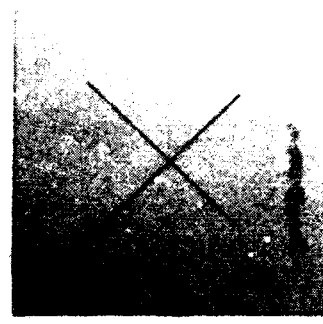
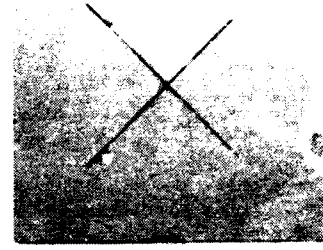
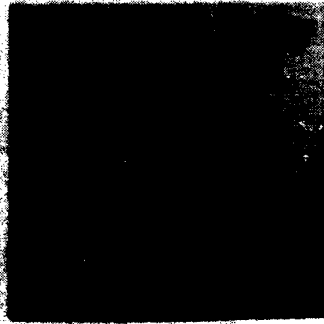
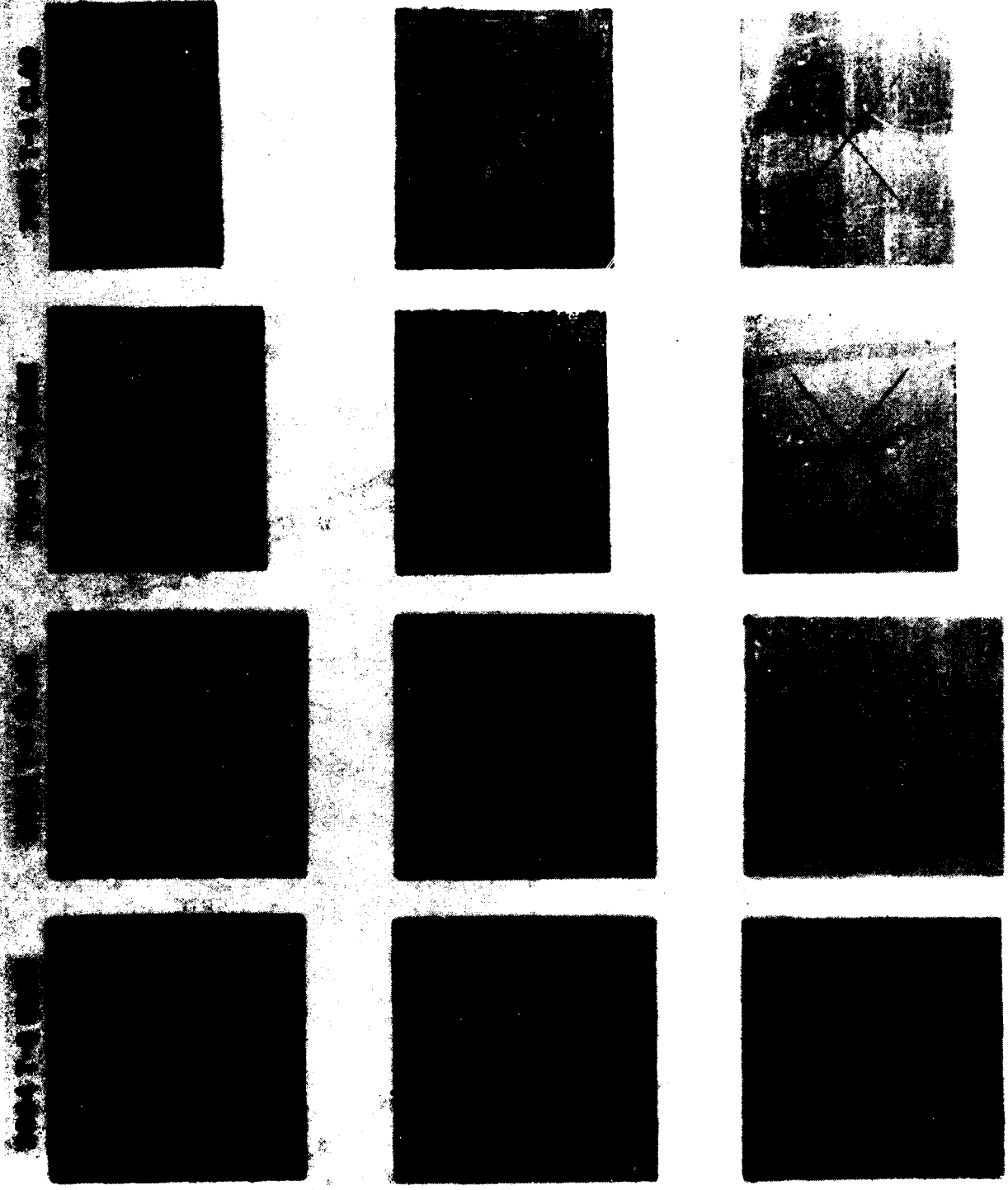


FIGURE 6: SULFURIC ACID ANODIZED-DICHROMATE SEALED SPECIMENS  
EXPOSED TO SULFUR DIOXIDE-SALT SPRAY FOR 500 HOURS.



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FIGURE 7: CHROMATE CONVERSION COATED SPECIMENS EXPOSED TO SULFUR DIOXIDE-SALT SPRAY FOR 500 HOURS.

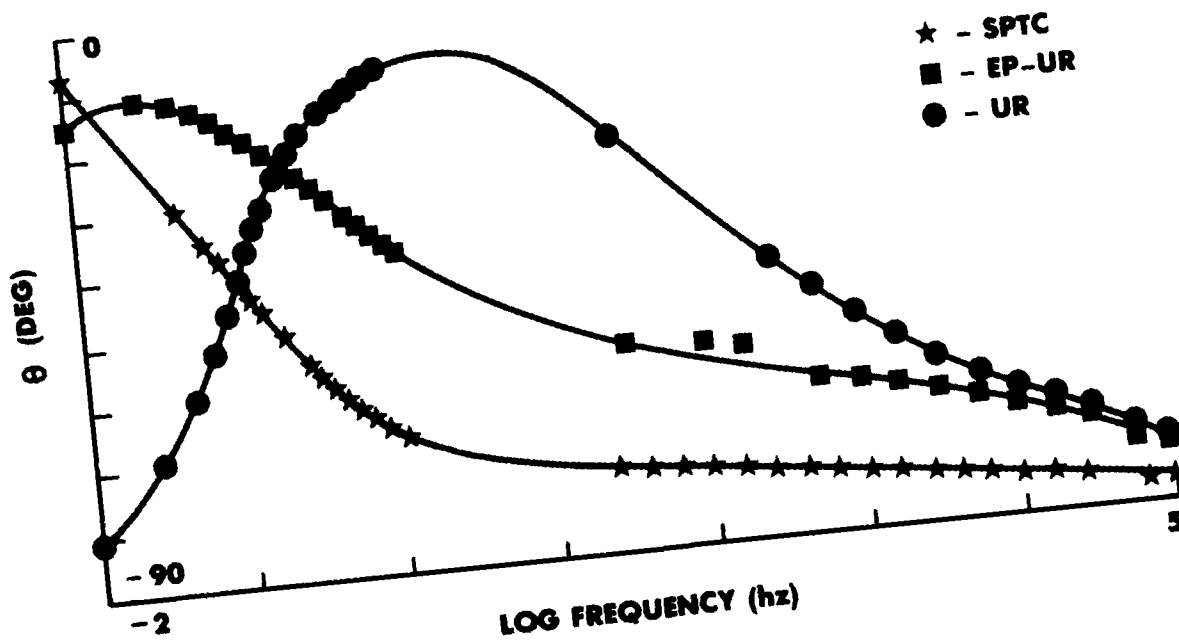
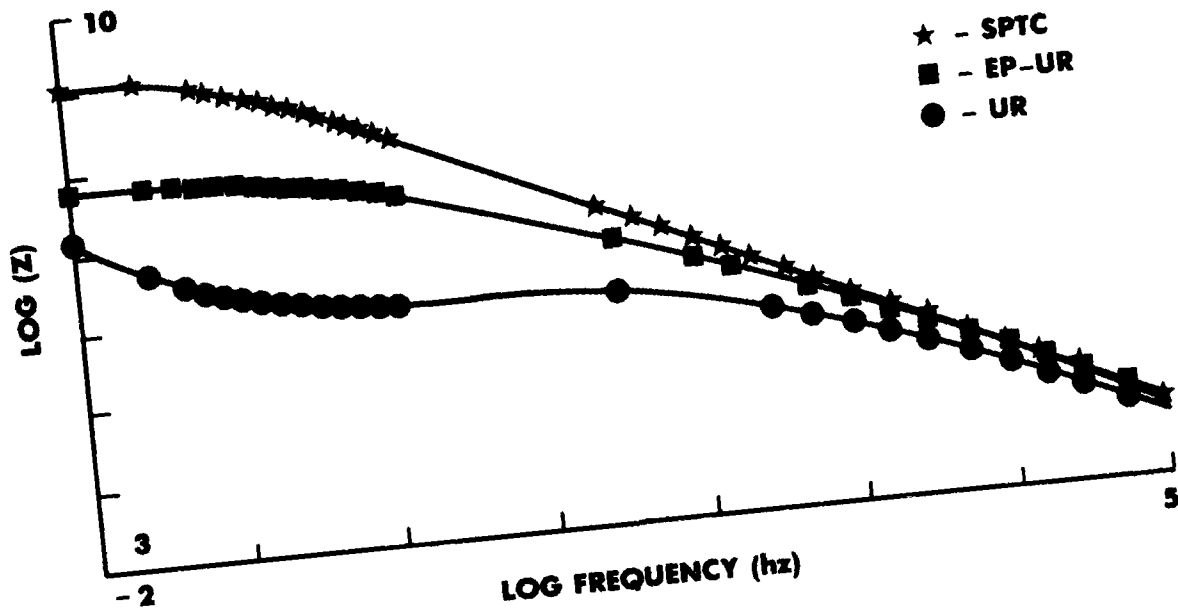


Figure 8. Bode plot of the three coating systems on the bare 7075-T6 aluminum alloy with the SAA/dichromate seal pretreatment after 24 hours.

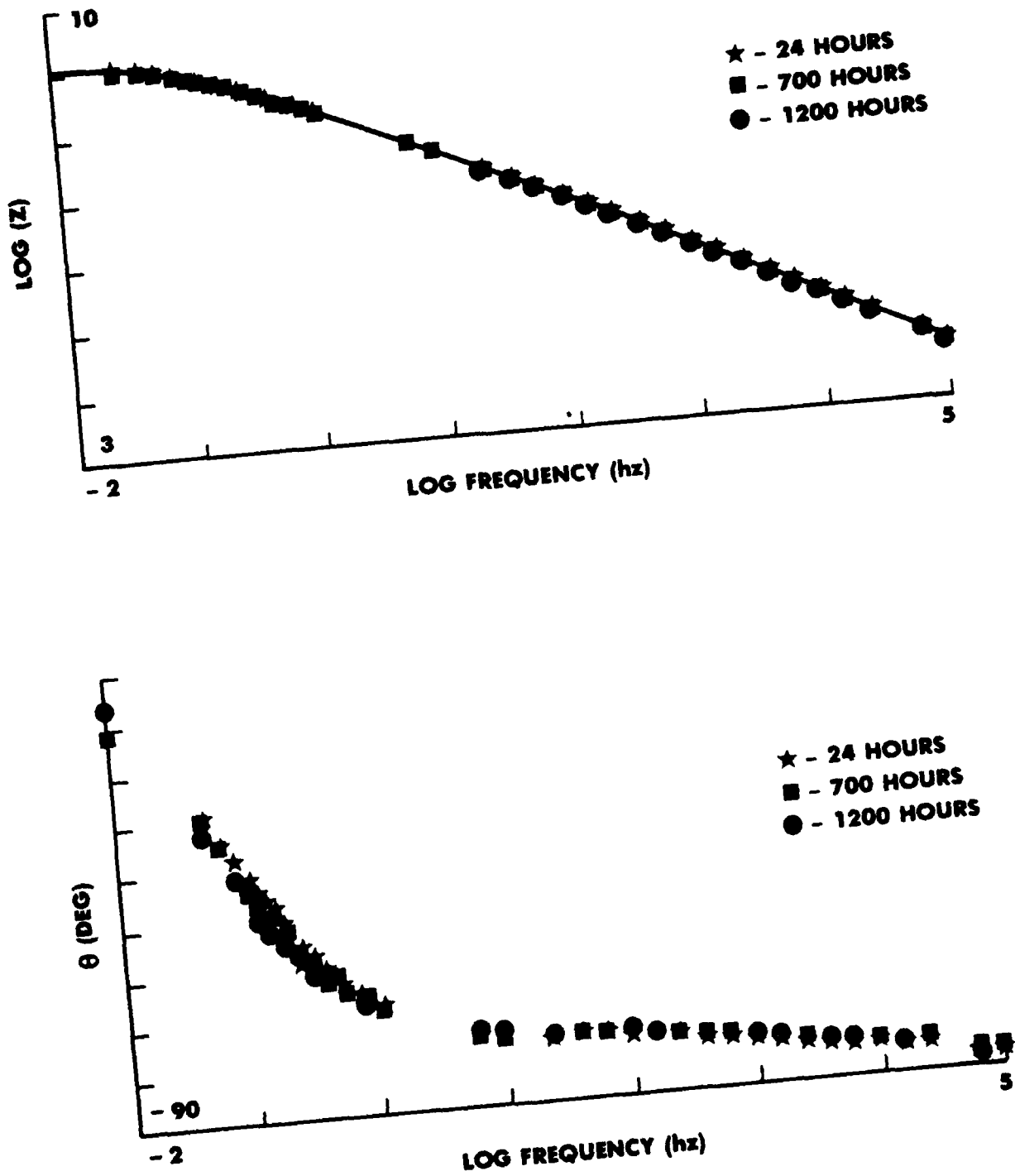


Figure 9. Bode plots for the SPTC system on the bare 7075-T6 aluminum alloy with the SAA/dichromate seal pretreatment at 24, 700 and 1200 hours.

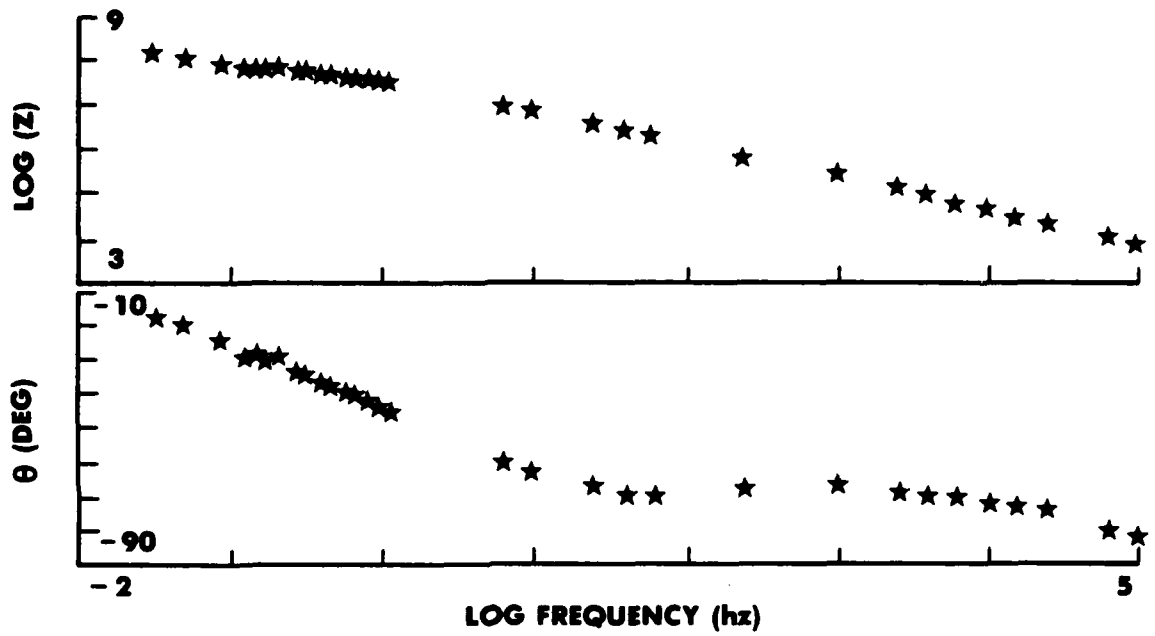


Figure 10. Bode plot of the EP-UR coating system on bare 7075-T6 aluminum with the chromate conversion coating pretreatment at 1200 hours.

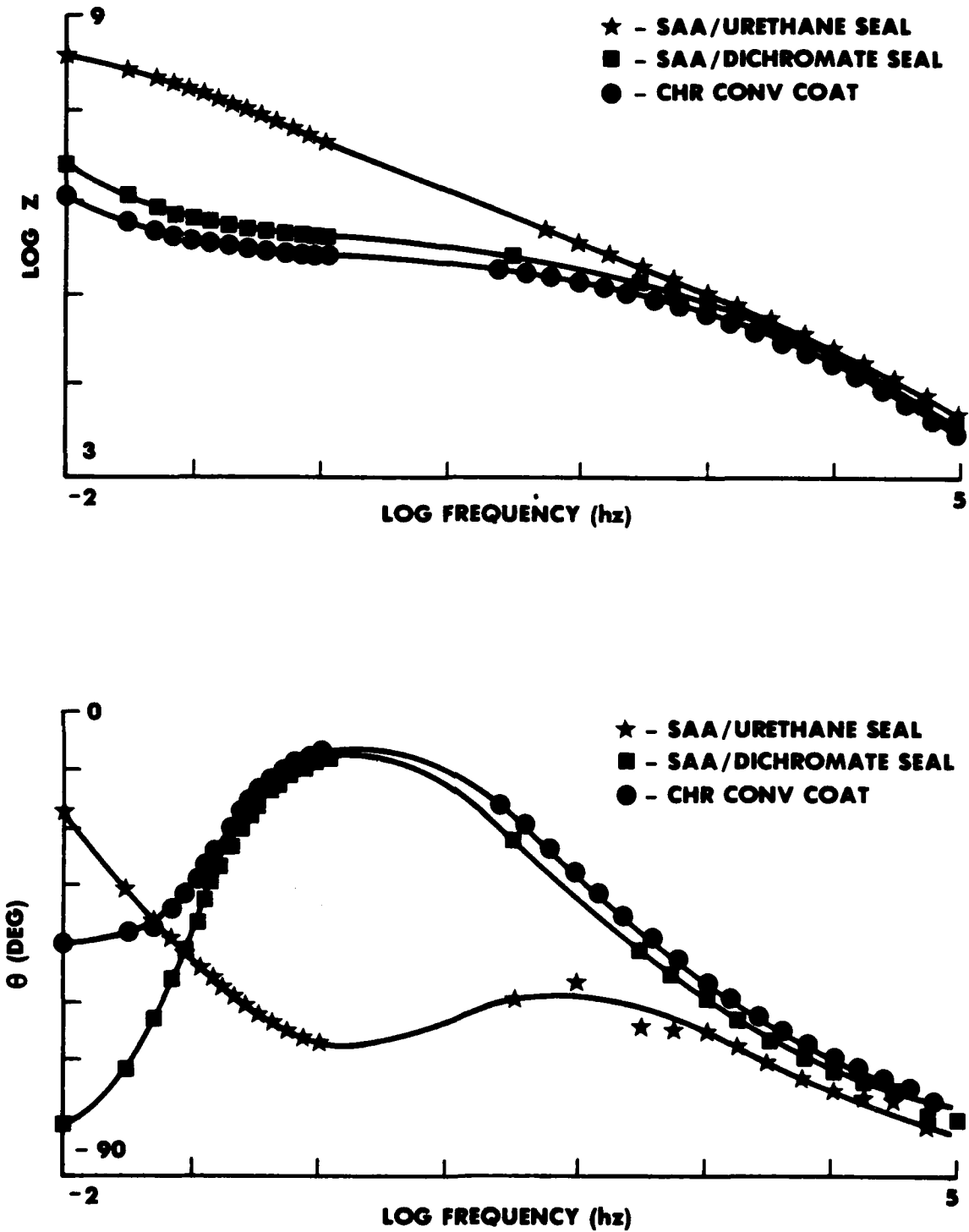


Figure 11. Bode plot of the UR coating on the three pretreatment systems after 24 hours.

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