



A review on Black coatings for Solar Energy Storing Systems

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Abstract: Solar energy, because of its abundance and everlasting nature, has established itself as one of the prime alternate sources of energy which could supplement and may eventually replace the fast depleting conventional sources of energy like fossil fuels and nuclear material. Black coatings are widely used in numerous applications e.g., decorative coatings, solar panels, optical instruments[1]. In this paper, an attempt has been made to review the different techniques of black coatings used in the conventional industrial methods and black coatings for solar applications are particularly presented and discussed.

Keywords : Black coatings, conversion coatings, absorptance, emittance.

Introduction

Black Electroplating

Process parameters for obtaining optimal solar selectivity using black nickel coatings on nickel have been discussed for NaCl solution[2]. Black nickel on SS303 have been discussed for nucleation and growth characteristics of electroplated layers of nickel[3]. Molybdenum black coatings on aluminium have been analysed for its electrical properties[4]. Mechanical properties of a trivalent chromium black coatings on cobalt has been optimized and exhibited better results in annealed conditions by Jeeva et al[5]. X-ray photoelectron spectroscopy (XPS) and AES techniques are used to explain the surface analysis of a black copper coatings under vacuum conditions[6].

Sharma et al discussed thermal behaviour of black anodic coatings on magnesium alloys and revealed that the thermal emittance of coatings increases with temperature[7]. Corrosion resistance of black chromium coatings under the influence of HCOOH have been discussed by Surviliene et al[8]. Ternary black Ni-Cu-Coelectrodeposited automobiles components are investigated by Jeeva et al for understanding its surface properties using XPS[9].

Black conversion coatings

Phosphating is the most widely used metal pre-treatment process for the surface treatment and finishing of ferrous and non-ferrous metals in the automobile, process and appliance industries due to its economy, speed of operation and ability to afford excellent corrosion resistance, wear resistance, adhesion and lubricative properties[10]. Zn-Fe MB 47 is exhibited high level corrosion protection coatings under salt spray analysis[11]. A method of forming a black conversion coating on surfaces of aluminum and its alloys has been discussed by Rodzewich[12]. Zinc plating using new single-dip black chromate conversion coating delivers better corrosion resistant[13]. Chemical and electrochemical treatments for black coatings on zinc and tin have been discussed by Gabe et al[14]. Gigand et al explained the mechanism of formation of chromate conversion coatings used

in the automobile companies for improving aesthetic aspect[15]. Zinc electroplated on aluminium surfaces using electrochemical conversion coating technique exhibited better optical properties[16].

Black inorganic anodized aluminium alloys are used after thermal cycling on 2XXX and 7XXX aluminium alloys for managing passive thermal control on spacecraft[17]. A dense and uniform black conversion coating on C1008 steel exhibited better corrosion resistance and higher efficiency of light-absorption[18]. Synergistic effects between chromate conversion coating and zinc-alloy deposit on corrosion resistance of steel was reported by Long *et al*[19].

Using TiAlON as source material for black decorative coatings improved wear resistance properties using PVD process[20]. Sharma *et al* revealed the substrate preparation and morphology of the zinc coatings on aluminium substrates [21]. The new phosphate black coating was developed by Jeeva *et al* [22] for improving the life time of machineries.

Solar selective coatings

Selective coatings are those which are having optical properties of reflectance, absorptance, transmittance and emittance spectrally dependent, in other words which vary significantly with wavelength so that the collection of thermal energy is correspondingly enhanced.

A selective absorber surface, for successful operational use in solar thermal systems, must possess as many of the following characteristics as possible: High solar absorptance (absorptivity >0.90), Low thermal emittance (emissivity <0.20), Large angle of acceptance, Long-term stability at desired operating temperatures and environmental conditions, Stability to (or recovery from) short-term overheating due to failure to extract energy from the collector, Stability at high temperatures during collector stagnation, Stability in the presence of moisture, Durability for the life of the collector, Applicability to given substrate materials, Reproducibility and Reasonable cost.

The various methods for preparation of selective coatings for solar thermal applications are: electroplating; chemical vapour deposition (CVD); vacuum evaporation; sputtering; anodization; chemical conversion; solution growth; spray; spray pyrolysis.

The advantages of Solchrome solar selective coatings are High absorptivity of $96\% \pm 2\%$, Lower emissivity of $12\% \pm 2\%$, Longer life, Higher operating efficiency, Higher thermal conductivity, Higher thermal efficiency, Suitable for heating fluids up to 85°C , Durability, More Kcal per money invested, State-of-the-art technology, High standard of quality, More energy generated per square meter installed and Low maintenance costs.

Solar selective layers on absorbers for solar collectors improve their thermal efficiency. A high solar absorptance a in the wavelength range of $0.3\text{-}2.0\ \mu\text{m}$ and a low thermal emittance e in the wavelength range of $2.0\text{-}20\ \mu\text{m}$ is needed. An ideal solar selective (solar absorber) coating must have high solar absorptance and low emittance[23]. Solar selective black chrome coatings are commonly used for decorative as well as useful applications because of their durability and excellent optical properties [24]. Though black chrome coatings deliver excellent optical properties, the use of toxic hexavalent chromium and requirement of high current densities are the major hitches in producing these coatings was reported by Dennis *et al* [25]. The use of electrolytic coatings such as black chromium or black nickel on highly infrared reflecting metals to produce selective absorbers for solar thermal energy conversion[26].

Electrolytic coatings such as black chromium or black nickel are widely used for efficient conversion of solar thermal energy. In combination with highly infrared reflecting substrates these films can provide absorber-reflector tandems with good selective properties was reported by Muehlratzer *et al*[27]. The optical performance can be significantly determined by both the microstructure and the metal content of the absorber film[28].

The black nickel solar absorber was developed by Tabor[29] over 40 years ago. It is produced by electroplating a metal base in an aqueous solution containing nickel- and zinc sulphates. A solar selective coating consisting of nickel, zinc, nickel- and zincsulphide is then obtained.

The cost and effective utilization of solar energy requires an efficient solar coating as having a high absorptance coefficient over the wavelength range of solar radiations (0.2–3 μm) and low emittance coefficient for long wavelength radiations[30].

Solar selective black chromium coating was electrodeposited on pre-treated electroformed nickel substrates from a hexavalent chromium containing bath. The composition of the film were investigated by Anandan *et al*[31] before and after annealing at 400 °C for different durations. This thermal treatment has been found to stabilize the coating which can be beneficially used for improvement in the solar selective properties of the coatings.

Nickel-based black coatings were electrodeposited onto both sides of a pre-treated stainless steel substrate from sulphate-based baths. Ammonium thiocyanate was used as the blackening agent. The individual effects of different alloying elements on the resultant magnetization behaviour of the deposits were reviewed by Ananth *et al*[32]. The influence of the plating current density (CD) was also explored. The magnetic properties of members of the family of nickel-based black coatings were extremely sensitive to bath formulations and operating variables. The magnetic properties of the electrodeposited Ni-Zn-Cd-Fe-S co-deposit was found to be fascinating.

Electrochemically deposited Cu–Ni black coatings on molybdenum substrate from ethylenediaminetetraacetic acid (EDTA) bath solution are shown to exhibit good optical properties ($\alpha=0.94$, $\epsilon=0.09$). X-ray initiated Auger electron spectroscopy (XAES) of Cu and Ni also agrees well with XPS investigations. X-ray initiated Auger electron spectroscopy (XAES) of Cu and Ni also agrees well with XPS investigations[33]. Black chrome (ChromeOnyx) was electrodeposited onto mechanically roughened, mechanically polished and electrochemically polished copper plates in order to investigate the influence of substrate surface preparation on the optical properties of the coating. The spectral reflectance of black chrome in the visible spectrum was found to be unaffected by the substrate surface preparations considered in this study. However, the near infrared reflectance was dependent upon the surface preparation. The results suggest that the variations in near-infrared absorptance are a consequence of differences in the structure and composition of the films resulting from substrate preparation[34].

Barrera *et al*[35] discussed the fractal characteristics of black molybdenum coatings on copper and to relate the fractal parameters to the optical properties. The results showed that optical properties, including absorptance and emittance, are decreasing functions of the Hurst and multifractality indices. This suggests that coating surfaces with high absorptance and emittance values are related to complex coating morphologies conformed within a non-linear structure.

The optical characterization of chromium black coatings prepared by electrodeposition at CENG (J. Spitz) were presented by Behaghel *et al*[36] and it exhibited hemispherical integrated values of a $\alpha = 0.97$ and $\epsilon = 0.1$. A conversion coating process is described for the preparation of nickel-black selective surfaces on zinc-plated steel, galvanized iron, zincated aluminium and Zinalume. With this process, it is possible to obtain surfaces with solar absorptances ranging from 0.90 to 0.94 and thermal emittances ranging from 0.08 to 0.15[37].

A low cost technique of spray pyrolysis has been described for the growth of black cobalt selective surfaces on commercially available aluminum and galvanized iron substrates were developed by Choudhury and his co-researchers[38]. Films on galvanized iron substrates gave best results ($\alpha=0.91$, $\epsilon_{100^\circ\text{C}}=0.12$) for film thickness $\sim 0.24 \mu\text{m}$. Cindrella *et al*[39] studied the effect of the additive (1,2,3-benzotriazole) on the coating parameters. A new complexing agent (ammonium acetate) other than thiocyanate usually reported for the development of selective black coatings is used in their system. The developed system produced a black coating in the current density range of 6–8 A/dm². Optimized coatings possessed a solar absorptance (α) of 0.96 and thermal emittance (ϵ) of 0.12.

Ebrahimiet al[40] introduced a technique to make an ultra-black surface by employing nanoporous anodized aluminum oxide as a template and deposition of nickel–phosphorus nanowires by the electroless process. The optical properties were compared with two other processes; a conventional black Ni–P deposition and a nickel electro-coloring process, on aluminum substrate. The results showed that ultra-black duplex coating possessed an absorption coefficient higher than 99%, while emission coefficient decreased about 6%

compared with simple black electroless Ni–P. Calculation of ξ factor indicated that a value of 5.1 proved that optical properties in the duplex coated sample had a significant improvement.

Black chrome absorber coatings have been developed by CVD-technique involving deposition of chrome hexacarbonyl. The deposition of the black chrome absorber films by CVD proceeds through pyrolysis of chrome hexacarbonyl at atmospheric pressure in the presence of oxygen. The black chrome absorber coatings demonstrate a significant solar absorptance (as deposited $\alpha > 0.95$) coupled with a high infrared reflectance (as deposited $\epsilon < 0.1$) which is necessary for efficient conversion of solar energy into heat. Coatings deposited on steel substrates have remained intact and substantially after 1000 h of lifetime testing in air at 600°C. The deposition at atmospheric pressure offering the potential of large-scale continuous flow-through manufacture. Substrates of such long pipes can be easily coated for solar farm plants[41].

Black chromium coatings were electrodeposited from a 1-butyl-3-methylimidazolium tetrafluoroborate ([BMIm][BF₄]) solution containing Cr(III) under potentiostatic control on copper substrates was developed by Eugénio *et al*[42]. Homogeneous black chromium films with spectrally selective optical properties were produced by applying a potential of –1.5 V for 1800 s, at an electrolyte temperature of 85 °C. The coatings consist of a mixture of chromium oxide/hydroxide and metallic chromium. Black chromium selective solar absorber coatings deposited by sputtering and electrodeposition have been studied by Holloway *et al*[43] after heat treating. Both sputter-deposited Al₂O₃–Cr and electrodeposited Cr₂O₃–Cr black chromium coatings degrade at temperatures below 400°C by oxidation of chromium to Cr₂O₃ in the coating.

Hosseini *et al*[44] compared the properties and microstructure of molybdenum black coatings, which are relevant to their use as solar selective absorbers. The short wavelength absorptance α_V (effectively for the visible rang) has been determined from diffuse and specular reflectance measurements while the IR emittance ϵ_{IR} has been determined by a calorimetric method. A new electrolyte has been proposed by John *et al*[45]for the deposition of black cobalt selective absorber coatings. These coatings are used in solar collectors for photothermal conversion of solar energy. Optical properties of the coating produced under the optimum conditions were found to be $\alpha = 0.96$ and $\epsilon = 0.12$. Thermal stability and corrosion resistance tests showed good durability of black cobalt selective coatings for high temperature applications.

An electrodeposited black nickel solar absorber coating has been produced cathodically from a low concentration bath by John[46]. Optical properties of the coating produced under the optimum conditions are found to be $\alpha = 0.96$ and $\epsilon = 0.11$. Thermal cycling and corrosion tests indicate the durability and stability of the coating suitable for solar thermal energy conversion systems. Multilayered chrome-free black selective surfaces for solar thermal energy conversion were produced by a low-cost sol–gel dip-coating method. After optimization of the multilayer design, a solar absorptance of 0.95 and a thermal emissivity of 0.12 at 100 °C have been achieved by Joly *et al*[47].

Copper cobalt manganese silicon oxides used as black selective coating for CSP tubes were investigated by Joly *et al*[48] different methods. ToF-SIMS and XPS depth profiling revealed that a three layered coating could be deposited on stainless steel substrates exhibiting 0.96 of solar absorption and 0.12 of thermal emissivity at 100 °C. Electrodeposited black nickel coatings plated from chloride baths on metallic substrates have high solar absorptance coefficients (> 0.92) and low thermal emittances (< 0.15). This coating is very stable under thermal implication and humid conditions[49].

An economically viable electroless process, different from the conventionally used chemical conversion methods, has been developed by Kumar *et al*[50] to deposit nickel black on galvanized iron. Coatings with integrated absorptance (α) lying between 0.90–0.93 and total emittance at 100° C = 0.073 to 0.1 and stable upto 200°C have been obtained. The coatings contain Ni and Zn particles embedded in a suitable matrix of NiO, Ni₂O₃ and Ni₇P₃. The coatings contain practically no sulphides and hence are less susceptible to humidity degradation processes. Black Chrome coatings were prepared on Ni, Fe, Cu and stainless-steel substrates by an electroplating technique. The optimum black chrome coating was made on Ni substrates using newly synthesized electrolyte by Lee *et al*[51] at a current density of 0.3–0.4 A/cm² for 20–30 s. Its optical properties were absorptance (α) of 0.90–0.96 and emittance (ϵ) of 0.25–0.30.

The effects of pulse current (PC) electrolysis during electrodeposition of black chrome oxide (Cr₂O₃) on the optical properties and surface microstructure of the solar selective coatings were investigated by Lee *et*

al[52]. This method enabled to achieve good optical properties ($\alpha=0.973$, $\epsilon=0.17$) for PC-applied initial (as-deposited) black chrome coatings. Lira-Cantú *et al* [53] worked on the electrochemical deposition of nanostructured nickel-based solar absorber coatings on stainless steel AISI type 316L. A sol-gel silica-based antireflection coating, from TEOS, was also applied to the solar surface by the dip-coating method. The best solar absorptance and thermal emittance values obtained on stainless steel substrates were 0.91 and 0.1, respectively.

A solar selective black nickel-cobalt coating which is more stable than the widely used black nickel (NiS-ZnS) has been formed on zincated and zinc-electroplated aluminium substrates by a conversion process were developed by Mehra *et al*[54]. Coatings with an integrated solar absorptance α in the range 0.90–0.94 and a room temperature emittance ϵ in the range 0.07–0.12 have been obtained. The effect of various deposition parameters on the optical properties, microstructure, the effect of thermal annealing in air at various temperatures on the optical properties and microstructure of the coatings has also been investigated by the above researchers.

The properties of black chromate conversion coatings on electrodeposited zinc and Zn-Co alloy have been investigated by Nikolova *et al*[55]. A possible method is described for the determination of the optical thickness of the coating. It has been established that the presence of certain additives, e.g. ethoxylated aliphatic alcohols C12 - C14 with 18 ethoxy groups and Na_2SiO_3 affect the absorption properties and thickness of the coatings. The inclusion of small amounts (up to 1 wt.%) of cobalt in the zinc sublayer strongly influence the parameters of the chromate coating. Black coatings formed on zinc attain values for the integral coefficient of solar absorption of $\alpha_s = 0.88$ –0.90 and on Zn-Co, $\alpha_s = 0.90$ –0.93.

Patel and Ianl[56] have investigated on solar-selective black zinc coatings through electrochemical treatment of zinc-electroplated aluminum substrates. Non-linear programming was used to optimize the anodizing parameters in terms of the solar absorptance and thermal emittance of the coatings. Optimum heat-treated (at 200°C for 200h) coatings with absorptances of 0.954 and emittances of 0.101 were produced with good reproducibility. Black nickel selective solar surfaces were produced from a nickel chloride bath with additions of zinc chloride, ammonium chloride, and sodium thiocyanate. Coatings with absorptances of 0.95 and emittances in the range of 0.24 to 0.26, after heat treatment at 200°C for 18 h, were produced[57].

Pettit[58] has discussed the accelerated aging of electrodeposited black chrome coatings deposited onto nickel substrates has been studied over the temperature range 350 to 450°C for coatings plated from four different bath compositions. It was found that the solar absorptance initially remains unchanged at values of 0.96–0.97 and then begins to gradually decrease with continued aging to values below 0.90. Investigations have been carried out by Pillai and Agarwal[59] on spectrally selective solar absorbing coatings of black chrome prepared by spraying an aqueous solution of CrO_3 on to chemically brightened aluminium substrates heated to 250–300°C. The advantages are good stability up to 250°C, low cost and the fact that the coatings can be used on collectors of any desired size. The coatings have high absorptivities in the solar spectral region ($\alpha_s \approx 0.93$) and low emissivities in the infra-red region ($\epsilon(100^\circ\text{C}) \approx 0.16$). Solar selective black nickel-cobalt plating on pre cleaned aluminum alloy substrates with nickel undercoat were investigated by Saxena *et al*[60]. The process provides high solar absorptance (0.948) and low thermal emittance (0.17), which is suitable for solar selective applications.

A new electrolyte has been proposed by Srinivasan *et al*[61] for the deposition of nickel-black selective coatings for use in flat-plate solar collectors. The coating exhibits better corrosion resistance than the well known black-nickel coatings (which contains nickel, zinc and sulphur) apart from having thermal stability upto 380°C. With no zinc in the deposit, this can be used in place of black nickel coatings with less corrosion problems.

The optical and structural properties of an electrolytic black cobalt absorber has been reported by Vitt[62]. Deposited on copper-coated steel substrates, this coating is used as a selective surface in the vacuum environment of the Philips evacuated receiver tubes. The material is shown to be well suited for the application even in high temperature evacuated tubular collectors, provided the annealing/degassing process is carried out under inert conditions.

Conclusion

A detailed literature discussion on black coatings has been summarized in this paper. The discussion covered the research findings of black coatings concerned both solar absorbance studies as well as mechanical properties as reported by several authors. This review could solve the literature gap for black coatings which are mainly used as solar absorbance studies and few researchers have claimed that industrial black coatings can be used as suitable candidate for improving the mechanical properties of metals besides its conventional applications.

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