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A SURVEY OF SELECTIVE SOLAR ABSORBERS AND THEIR LIMITATIONS (*)

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Abstract.—A number of materials and techniques have been used to form selective solar absorber coatings. Each of the coating processes and the coatings formed has its own limitations which are generally poorly defined. The poor state of knowledge on the limitations of the coating processes and the resulting coatings poses a problem for the component design and the materials engineer. This paper reviews the state of knowledge of some of the coating processes and materials being used and of potential usefulness in mid-temperature solar-thermal collectors. A principle point that is made is the lack of production experience and knowledge of the degradation modes for most of the coating technologies and coating material.

1. Introduction.—Solar radiation is a low intensity energy source whose utilization requires effective collection and retention to be economically feasible. Operational temperature ranges of solar collector systems may be categorized as: low-temperature (T < 100°C), mid-temperature (100 < T < 400°C), and high-temperature (T > 400°C). To obtain mid and high operational temperatures, some degree of concentration of the incident radiation is required /1/. In the mid-temperature range, the concentration (ratio of aperture area to receiver area) will generally be from 5 to 50 while in the high-temperature collectors the concentration may exceed 1000.

Collector designs utilize a receiver surface to absorb the incident solar radiation /1/. In the "coating receiver", the radiation is absorbed in a coating or surface layer and the heat is conducted through the container wall to the heat transfer media.

In the "container receiver", the radiation is absorbed within the container walls while the "fluid receiver", the radiation is absorbed in

(*) This work is supported by the U.S. Department of Energy under Contract DE-AC04-76-DP00789.
(**) A U.S. DOE facility.
the heat transfer fluid. In each of these designs, the optical properties of the construction materials must be optimized for maximum absorption efficiency and minimum radiation losses. The most common design is that of the coating receiver but the fluid receiver design is of interest for low-temperature collectors where radiation losses are minimal. In each of these collector systems, a stagnation temperature, which is much higher than the operational temperature, may be attained when the heat transfer fluid is not flowing.

In most receiver designs, the radiation absorbing surface is protected by a transparent cover (glazing) to reduce convective heat losses. In the most advanced designs, this cover provides a means to control the environment to which the absorber surface is exposed. In less advanced designs, the glazing may serve to trap undesirable elements such as moisture or outgassing products and create a harsh environment for the absorber surface [2]. The environments to which absorber surfaces may be subjected vary greatly but include: 1) greater than operational temperatures, 2) thermal shock and fatigue, 3) corrosive and oxidizing agents, 4) ultraviolet radiation, 5) moisture, 6) preassembly and assembly contamination and damage and 7) particle abrasion and accumulation (unprotected surfaces). Economics will often determine to what extent the environment will be controlled.

In any collector design, an effort is made to maximize the collector efficiency by minimizing reflectance losses from reflectors and glazing, minimizing absorption losses in the glazing and to maximize the absorption of solar radiation at the absorber surface. For low-temperature applications, the thermal radiation losses may not be important because of the low operational temperature while for high-temperature applications radiation may not be important because of the low receiver area. In the mid-temperature range, thermal radiation losses may be appreciable though for some "cavity receiver" designs much of the emitted thermal radiation may be trapped and absorbed. For applications where thermal losses need to be minimized, an absorber surface which has a high solar absorptance (αₚ) and a low thermal emittance at the operational temperature (εₚ) is desirable. Such a material is called a selective absorber [1, 3, 4, 5]. Typically, the designer will strive for an αₚ ≈ 0.95 and an εₚ as low as possible for the high αₚ (typically εₚ < 0.2).

Figure 1 shows the solar radiation spectrum external to the earth's atmosphere and after passing through two standard air masses (AM 2).
Fig. 1.- Solar spectrum external to the earth's atmosphere (AMO) and after passing through two standard air masses (AM2). Black body radiation spectra for several temperatures and a table of radiant energy contained in each spectrum.

The human eye is only sensitive to about 40% of the energy contained in the solar spectrum. Figure 1 also shows the radiation spectra from black bodies at selected temperatures. The table with the figure indicates the amount of energy contained in each of the radiation spectra. The total radiation emitted from a surface is proportional to the surface area, the emittance and the surface temperature to the fourth power.

In order for a solar collector to have a high efficiency and maintain a high efficiency over its operational life, it is necessary that the structure and degradation modes of the various components be understood. The selective solar absorber surface with its exacting optical properties is one of the areas most sensitive to degradation. Unfortunately, the degradation mechanisms of these surfaces is poorly understood.

2. Selective solar absorbers.- Selective solar absorbers may be fabricated from a number of materials and material combinations by a variety of techniques /4, 6/. Factors which determine which absorbers will be used for a specific application include: 1) absorptance and emittance properties, 2) stability in the operational environment, 3) ability to mass produce the absorber, 4) economics of fabrication and installation, and 5) designer bias due to lack of valid test data and performance ex-
perience at the present time for many applications.

![Diagram of a typical selective absorber coating](image)

**Fig. 2.** Typical selective absorber coating containing and absorbing layer on a low emittance substrate with a radiation trapping surface.

Figure 2 shows one type of selective solar absorbing surface which is generally formed by some coating or reaction technique /7/. The absorber consists of an anti-reflecting surface which may be produced by having a low index of refraction material in contact with the gas or vacuum environment or by having a surface morphology which gives radiation trapping. The transmitted radiation is absorbed in a layer having a high extinction coefficient for the solar radiation spectrum and which has a low volume emittance for thermal radiation. This absorbing layer is formed on a layer which has a low thermal emittance and provides corrosion protection for the underlying structure where necessary. This selective absorber coating configuration should have a high thermal conductivity to keep the coating surface temperatures as low as possible.

The low thermal emittance underlayer surface is generally a metal such as Ni, Mo, Cu or Al which has a thermal emittance of about 0.1 at 300°C /8/. These metal coatings may be fabricated by a number of techniques such as electroplating, sputter deposition, vacuum evaporation, or may be the structural material of the receiver. For non-corrosive environments, this underlayer may be very thin but where the underlayer must provide corrosion protection for the structure, the layer must have an appreciable thickness which will depend on the service conditions. For example, electroplated nickel on mild steel should be greater than 0.00127 cm (0.5 mil) when used in an air/moisture environment in order to provide corrosion protection in our test conditions.

The absorbing layer of the coating may be comprised of a metal-dielectric cermet, a metal-dielectric layered configuration or a semiconductor having an optical absorption edge near 2000nm. In all of these systems, it is desirable to keep the thickness of the absorber material as small as possible since the emittance will increase with thickness.
Semiconductor absorbers which have been fabricated include silicon, germanium, and lead sulfide. Since these semiconductors all have high refractive indices, antireflection surfaces must be used to prevent excessive reflectance losses. Amorphous silicon prepared by plasma decomposition of silane has good absorption characteristics, though when hydrogen is incorporated into the deposit the absorption edge is shifted further from the near infrared /9/. Chemically vapour deposited (CVD) silicon has good absorption properties but at temperatures above 500°C, the α-silicon structure is unstable and the optical properties will change. The addition of carbon to the CVD-Si stabilizes the structure to > 700°C /10/. Evaporated germanium is a good selective absorber /11/ but little has been reported on its thermal and chemical stability. Evaporated lead sulfide is also a good selective absorber /11, 12/ but converts to the sulfate when exposed to oxygen in the presence of ultraviolet radiation /11/.

Layered structures may be fabricated by evaporation, sputter deposition or electroplating. One of the most successful such coatings is the Al₂O₃-Mo-Al₂O₃ (AMA) interference type selective absorber coating /13, 14, 15/. The topmost Al₂O₃ coating (80 nm) acts as an antireflection coating while the Mo (or MoOₓ) (10 nm) and the underlying Al₂O₃ (30 nm) act as the absorber. This layered structure deposited on an Mo underlayer has yielded an αₚ = 0.95 and ε(20°C) = 0.07. This type of structure may degrade by oxidation of the Mo absorption layer or by interdiffusion of the layers.

Cermet layers have been the most widely studied group of absorption layers. Cermets may be fabricated by electroplating, anodizing-impregnating, inert or reactive sputter deposition and vacuum evaporation. Electroplated "black chrome" is a widely used selective absorber for the lower mid-temperature ( < 300°C) range /16, 17, 18, 19/. This coating is comprised of Cr particles in a CrₓOᵧ matrix in a rather open structured coating /16, 17/. The chromium content increases toward the substrate and the principle degradation modes seem to be internal oxidation and/or diffusion from the electroplated underlay. The black chrome absorber is easily prepared but suffers thermal instability above 250°C without control of the composition of the electroplating bath /20, 21/. Table I shows test results for samples prepared with differing bath compositions /21/. By using a low concentration of Cr⁺³ in the bath, a coating which is stable to 300°C in air can be produced. For the low Cr⁺³ baths, the deposition rate decreases with plating time. Thermal degradation of the black chrome coating is first evidenced by a decrease in absorbance in the 1000-1500 nm range as is shown in figure 3.
Table I.- Thermal Aging of Electrodeposited Black Chrome /20/

<table>
<thead>
<tr>
<th>Plating Conditions</th>
<th>Test Temp.</th>
<th>Test Time (hr)</th>
<th>$a_s$</th>
<th>$e(100^\circ C)$</th>
<th>$e(300^\circ C)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>16 g/l Cr$^{3+}$</td>
<td>As Plated</td>
<td>250$^\circ C$</td>
<td>0</td>
<td>0.95</td>
<td>0.09</td>
</tr>
<tr>
<td>3.5 min.</td>
<td>350$^\circ C$</td>
<td>64</td>
<td>0.93</td>
<td>0.08</td>
<td>0.14</td>
</tr>
<tr>
<td></td>
<td></td>
<td>64</td>
<td>0.89</td>
<td>0.07</td>
<td>0.12</td>
</tr>
<tr>
<td>12 g/l Cr$^{3+}$</td>
<td>As Plated</td>
<td>250$^\circ C$</td>
<td>157</td>
<td>0.97</td>
<td>0.21</td>
</tr>
<tr>
<td>3.5 min.</td>
<td>350$^\circ C$</td>
<td>1849</td>
<td>0.96</td>
<td>0.17</td>
<td>0.22</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3908</td>
<td>0.95</td>
<td>0.16</td>
<td>0.22</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0</td>
<td>0.97</td>
<td>0.21</td>
<td>0.31</td>
</tr>
<tr>
<td></td>
<td>400$^\circ C$</td>
<td>157</td>
<td>0.96</td>
<td>0.17</td>
<td>0.23</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1596</td>
<td>0.88</td>
<td>0.14</td>
<td>0.18</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3651</td>
<td>0.86</td>
<td>0.14</td>
<td>0.10</td>
</tr>
<tr>
<td>8 g/l Cr$^{3+}$</td>
<td>As Plated</td>
<td>350$^\circ C$</td>
<td>157</td>
<td>0.97</td>
<td>0.29</td>
</tr>
<tr>
<td>5 min.</td>
<td></td>
<td>1849</td>
<td>0.98</td>
<td>0.22</td>
<td>0.40</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3908</td>
<td>0.98</td>
<td>0.23</td>
<td>0.32</td>
</tr>
<tr>
<td></td>
<td>400$^\circ C$</td>
<td>0</td>
<td>0.97</td>
<td>0.29</td>
<td>0.31</td>
</tr>
<tr>
<td></td>
<td></td>
<td>157</td>
<td>0.97</td>
<td>0.21</td>
<td>0.29</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1595</td>
<td>0.95</td>
<td>0.18</td>
<td>0.24</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3650</td>
<td>0.94</td>
<td>0.17</td>
<td>0.23</td>
</tr>
</tbody>
</table>

Fig. 3.- Typical reflectance spectrum for thermally unstable electrodeposited black chrome showing increase reflectance (decreased absorption) in the 1000-1500 nm range on thermal degradation.

The more stable coatings (8 g/l Cr$^{3+}$) seem to be comprised of larger particles than the less stable coatings (16 g/l Cr$^{3+}$).

Inert and reactive sputtering have been extensively used to prepare cermet coatings. These include MgO-Au /22, 23/, Cr$_2$O$_3$-Cr /24/, Al$_2$O$_3$-Cr /25/, Al$_2$O$_3$-Mo /25/, Al$_2$O$_3$-Pt /26/ and ZrC$_x$ /27, 29/. The coatings appear to be very small metallic phase particles (5-10 nm) in an amorphous ceramic matrix.
Table II summarizes a number of selected systems as to the fabrication technique and the degradation temperature and mode where known.

Table II.— Selected Selective Solar Absorbers

<table>
<thead>
<tr>
<th>Material Coating/Substrate</th>
<th>Fabrication Process</th>
<th>$a_2/\epsilon$ ($^\circ$C)</th>
<th>Operating Temp.</th>
<th>Degradation Mode</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Au-MgO/Mo</td>
<td>Sputtering</td>
<td>0.93/0.69 (20)</td>
<td>400</td>
<td>300 (? )</td>
<td>22,23</td>
</tr>
<tr>
<td>Cr-Cr$_2$O$_3$/SiO$_2$</td>
<td>Sputtering</td>
<td>0.92/0.68 (20)</td>
<td>&gt;300</td>
<td>000</td>
<td>24</td>
</tr>
<tr>
<td>Pt-Al$_2$O$_3$/SiO$_2$</td>
<td>Sputtering</td>
<td>0.94/0.07 (20)</td>
<td>---</td>
<td>500</td>
<td>26</td>
</tr>
<tr>
<td>ZrC$_x$/Zr</td>
<td>Sputtering</td>
<td>0.90/0.05 (20)</td>
<td>---</td>
<td>800</td>
<td>27-29</td>
</tr>
<tr>
<td>Fe-SiO$_2$/Cu</td>
<td>Sputtering</td>
<td>0.90/0.04 (20)</td>
<td>---</td>
<td>---</td>
<td>31,32</td>
</tr>
<tr>
<td>Al$_2$O$_3$-Mo-Al$_2$O$_3$/Mo</td>
<td>Sputtering</td>
<td>0.94/0.08 (20)</td>
<td>450</td>
<td>600</td>
<td>Diffusion 13,14</td>
</tr>
<tr>
<td>Al$_2$O$_3$-ZrC$_x$/Ag</td>
<td>Sputtering</td>
<td>0.91/0.05 (325)</td>
<td>175</td>
<td>700</td>
<td>33</td>
</tr>
<tr>
<td>Cr-Cr$_2$O$_3$/Ni</td>
<td>Electrodep.</td>
<td>0.94/0.15 (20)</td>
<td>300</td>
<td>---</td>
<td>Ox/Diffusion 16-21,34,35, 43,48,49</td>
</tr>
<tr>
<td>Co$_x$/Ni</td>
<td>Electrodep./Thermal</td>
<td>0.93/0.1 (100)</td>
<td>400</td>
<td>---</td>
<td>Morp. Change 36,37</td>
</tr>
<tr>
<td>AR/aSi(+C)/MoX</td>
<td>CVD</td>
<td>0.91/-</td>
<td>---</td>
<td>&gt;700</td>
<td>Crystal 10</td>
</tr>
<tr>
<td>Pb/S/Al</td>
<td>Evaporation</td>
<td>0.93/0.21</td>
<td>&gt;100</td>
<td>&gt;120</td>
<td>Oxidation (UV) 11,12</td>
</tr>
<tr>
<td>AR/Molybdenum</td>
<td>CVD</td>
<td>0.82/0.68 (20)</td>
<td>---</td>
<td>---</td>
<td>39</td>
</tr>
<tr>
<td>Co$_2$O$_4$/Ag/SS(0x)</td>
<td>Spray Pyro.</td>
<td>0.91/0.20 (20)</td>
<td>&gt;650</td>
<td>---</td>
<td>50</td>
</tr>
<tr>
<td>Ni-Al$_2$O$_3$/Al</td>
<td>Anodization</td>
<td>0.95/0.15 (20)</td>
<td>---</td>
<td>---</td>
<td>51</td>
</tr>
</tbody>
</table>

SS = Stainless Steel  
AR = Antirefection Coating  
Ox = Oxidation

Antireflection coatings may be deliberately deposited using materials with low indices of refraction and specific thicknesses. Antireflection or radiation trapping surfaces may be in the form of: 1) dendrites, 2) cavities, 3) powders, or 4) etch features. The spatial dimensions of the features must be on the order of the wavelength of the solar radiation (500 nm). The trapping morphology may be formed in a number of ways including: 1) inherent growth structures, 2) chemical etching, 3) sputter etching, or 4) deposition of fine particles.

Growth structures such as dendritic or columns form naturally under some deposition conditions or during chemical conversion of surfaces. Such features may be found on electroless nickel /38/ CVD tungsten /40/ and molybdenum /39/, sputter deposited Ge and Si (accentuated by chemical etching) /41/, and many other systems. Chemical etching may be used to roughen a semiconductor surface or to selectively etch a co-deposited phase (Al from sputter deposited ZrC + Al)/33/. Chemical etching has also been used to convert a nickel surface into a highly absorbing surface /38/. Sputter etching of copper in a carbon atmosphere has been used to generate a highly absorbing surface /42/. The deposition of fine particles from the liquid or gas phase has been used to
produce highly absorbing surfaces of metals /23/ and germanium /11/. The porous nature of these deposits gives high surface temperatures and may result in sintering of the coatings or increased thermal losses through radiation.

3. Degradation modes.- Generally, the degradation modes for the various coatings are poorly understood and are dependent on the test environment and conditions. Since generally very small amounts of material are involved in any changes, it is generally difficult to unambiguously specify the degradation mode. Degradation can be promoted by thermal processes (including diffusion) and chemical processes. Both processes are temperature, time, environment and material dependent.

The most obvious degradation mode is loss of adhesion of the coating generally due to poor initial adhesion or interfacial corrosion or reaction. Pinhole corrosion leads to degradation of the coating surrounding the pinhole region.

The optical properties of the coatings generally degrade due to loss of absorptance and not to an increase in thermal emittance. Indeed there often seems to be a decrease in emittance with a decrease in absorptance. The loss of absorptance may be caused by changes in:

- surface morphology
- chemical or phase composition
- phase distribution or size

For elevated temperatures, diffusion from the underlay coating often seems to be a problem. Most electroplating baths have a number of organic and inorganic additives which become incorporated into the deposit and are subsequently able to diffuse into the coating along with the metal. This diffusion will be promoted by an "open structure" of the absorbing coating which allows surface or grain boundary diffusion.

Chemical reaction rates will also be increased by having an "open structure" to the absorbing coating. An oxidizing environment will allow the conversion of metallic phases to oxides thus reducing the absorptance of cermet or layer type absorbers. The presence of an oxidizing environment enhances diffusion of oxidizable metals from the interface to the surface /44/. The presence of water in the environment probably increases the reaction rate.

The most stable system for oxidizing environments seem to be ones which use refractory metal underlayers (Mo) with oxidation resistant (Pt, Au) metal particles in an oxide matrix /26/. It has been suggested that a diffusion barrier on the metal underlayer may provide increased stability /14/.

4. Fabrication Processes.- For several years, Sandia National Laboratories has been actively engaged in using electrodeposited black chrome
for a mid-temperature absorber surface in a line concentrator configuration. Initial development and test configurations were coated in the laboratory but production has been done at several commercial electroplating firms. The desired coatings should have a solar absorptance of 0.95 and an emittance of \(< 0.20\) at the operating temperature \(\sim 320^\circ C\). The coatings are not subjected to higher stagnation temperatures since fluid transfer failure results in turning the concentrator through to the store position. Recent results with this concentrator using silvered, chemically strengthened glass reflectors and the improved black chrome absorber coating has given a 60% collection efficiency at 320°C.

In the laboratory, a number of important deposition variables have been identified as to performance and stability of the black chrome coating. The initial bath which was used was the Harshaw Chromonyx® formulation developed for decorative black coatings. It was found that careful control of the total charge passed yielded an optimum solar absorptance with a minimal emittance /16/. The current density for a good coating is in the range of 150-200 A/ft² at 18-28°C for 4-1/2 to 6 minutes giving a coating thickness of about 2000 Å at an anode-to-cathode ratio of 2-3/1. These conditions may vary with bath composition.

Thermal stability studies have shown that the Cr³⁺ concentration in the Chromonyx® bath has an important effect on the subsequent thermal stability of the coating as shown in table I /20/. In order to obtain a thermally stable coating for use above 250°C, it was found necessary to control the Cr³⁺ concentration very carefully. The black chrome coating is routinely deposited on an electroplated nickel underlayer and if the nickel is not plated immediately with the black chrome, the surface must be activated by an HCl etch. On occasion, the nickel is apparently not activated completely and the resulting black chrome coating is not thermally stable, possibly due to poor nucleation of the electrodeposited black chrome on the nickel surface.

From our laboratory results, it is apparent that to obtain high quality black chrome coatings it is important that there be:

1. Continuous bath analysis
2. Optical monitoring of parts during fabrication

When the black chrome process was transferred to commercial electroplating firms, a number of problems surfaced. Generally, electroplaters are not used to the stringent deposition requirements needed to obtain the high quality optical coatings needed. In addition, these firms do not have the necessary manpower, expertise or equipment necessary to do the required chemical analysis on the bath nor measure the desired
optical properties of the coatings. It was found necessary for Sandia personnel to be present during all production runs of our product in order to make optical measurements on the production coatings. It has been our experience that the electroplating baths used to produce thermally stable coatings will deteriorate over a period of time. This may be in part due to variables such as contaminants which we have not identified and thus do not control. In addition, any change in substrate geometry results in the process having to be adjusted to achieve optimum results.

A high volume optical coating deposition process which has proven to be reliable is that of sputter deposition which is widely used to produce coated architectural glass [45]. Several groups are using sputter deposition to deposit selective solar absorbing coatings [32, 46]. A cost analysis of this process would seem to indicate that the process is cost competitive with electroplating in high volumes [31].

Other processes such as anodization/impregnation [51], thermal oxidation, spray pyrolysis [50], chemical vapour deposition [47], or other coating techniques may prove to be cost competitive and feasible for commercial scale-up but these techniques have yet to prove themselves in a mass production environment.

5. Summary.- A number of selective absorber coating systems with high solar absorptance exist which may be used in the mid-temperature range. Some of the systems are more chemically and thermally stable than others. Unfortunately, there are large gaps in the stability data for a large number of the systems.

In an inert environment, the principle degradation mechanisms are interdiffusion between the layers or phases and changes in surface morphology. These degradation mechanisms would be minimized by using refractory metals and compounds for the absorbing layer and using refractory materials or diffusion barriers for the underlayer.

For use in a reactive environment, the choice of materials is much more restrictive since internal chemical reactions can change phase compositions and interfacial reactions can lead to loss of adhesion.

For a coating process to be useful, it is necessary to determine what parameters influence the performance of the coating and the limits to these parameters. This "process sensitivity" has a direct influence on the production process controls necessary to produce a good product. Again, much of this type of information is not available on a number of coating systems.

Our experience with electroplated black chrome has been rather disappointing. Electroplating should be a low cost deposition process but the extensive bath analysis and optical monitoring necessary to
produce a thermally stable produce for use to 320°C has increased our cost significantly. It seems to be rather difficult to obtain the necessary production process controls. It should be noted that the black chrome electroplate is quite satisfactory for use at temperatures below 250°C.

In view of the increased cost and complexity necessary to extend the thermal stability range of the present electrodeposited black chrome from 250°C to 350°C, it would seem that some other deposition technique might provide a significant improvement over electroplating for coatings to be used above 250°C.

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