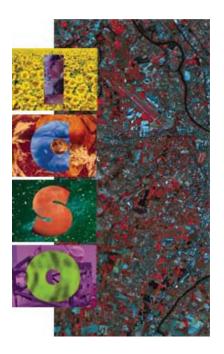
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DESIGN AND MANUFACTURE OF HIGH ABSORPTION METAL DIELECTRIC COATINGS FOR THE REDUCTION OF STRAYLIGHT

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ABSTRACT - This paper describes the design and manufacture of broadband metal dielectric absorbers. First, we give some design principles to obtain achromatic absorption properties. Then, we describe a new method to determine the complex refractive index of metallic layers. A graded index model is developed to take account of the evolution of the film packing density. Manufacturing is detailed in the last section. Absorption levels higher than 99.9% have been measured over the visible range.

1 – INTRODUCTION

Broad band light absorbers are key components to suppress stray light. Such components should be able to absorb light whatever the wavelength, the angle of incidence and the state of polarization. The most common and versatile solution to do so is certainly the use of black paints. This technique permits to cover easily large areas of various shapes with fairly high efficiency: Just a few percent of the incident light is not absorbed but fully scattered. If scattering is not appropriate, a similar result can be achieved using black glasses. In that case, the few percents of non-absorbed light are specularly reflected. However, the absorption level provided by these two solutions may be too low for some critical applications or in other words, the amount of scattered or reflected light is still too high. In the case of black glasses, a classical broad band antireflection multi-layer coating permits to increase the absorption level higher than 0.99. In the case of black paints, Giovanini and Amra also showed how the use of a dielectric coating, deposited on such a rough surface, permits to reach similar performance, as shown in figure 1 [1]. For both solutions, the absorption takes place inside the substrate and since these substrates are made of weakly absorbing material, black glass or black paint, these substrates should be rather thick to absorb all the light that penetrates them, at least one hundred micrometers. However, for a few special micrometric applications, the thickness is a critical parameter. We can cite for example the case of some absorbing diaphragms with very sharp edges. These diaphragms should be black coated but the sharpness of the edge is not compatible with a painting thickness [2]. We can also mention micro-baffling applications, as it is the case for the multi-spectral detector developed for SPOT [3]. In that particular case, absorbing coatings should be deposited between the CCD lines, and the thickness of these coatings should not exceed a few microns with an absorption level greater than 0.99. For these particular applications, we developed an third solution: metal-dielectric multi-layer absorbers.

The main idea is to use a metallic material to absorb light. Considering the high extinction coefficient of these materials, total absorption can be achieved within a few hundred nanometers. However, metallic materials also induce high reflectance that must be cancelled using an

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appropriate antireflection coating. The best design for this coating, which will be discussed in the next section, consists in a metal-dielectric antireflection coating, which requires very thin metallic layers. Because of these layers, we need an accurate index determination of metallic thin films to obtain optimum efficiency. To answer this problem, we developed new index determination methods we will describe briefly before considering the manufacturing of the components in the last section.

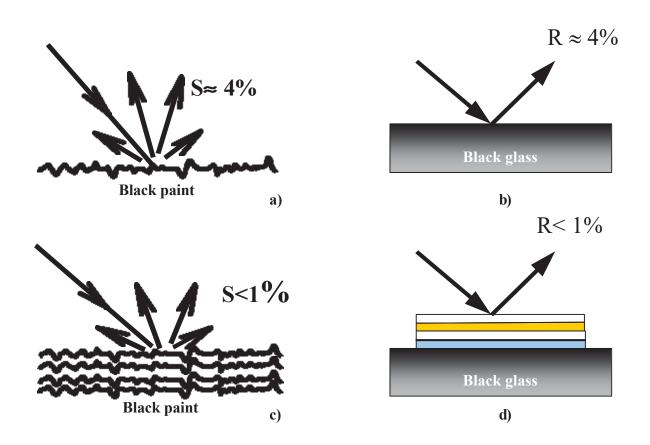


Fig. 1: Black paints and black glasses are common solutions to obtain achromatic absorption properties. The non absorbed light may be scattered of reflected (a and b). In both cases, the use of a dielectric multi-layer permits to increase the absorption (c and d).

2- DESIGN OF BROAD-BAND METAL-DIELECTRIC COATINGS

The easiest way to design an efficient broadband metal-dielectric coating is surely the use of a numerical optimization procedure [4]. As a consequence, we will mainly discuss in this section some general points such as the choice of materials. As said in the introduction, a metallic layer is used to absorb light. This layer is of course deposited on the component that must be black coated. This first layer is thick enough to avoid any transmitted light. As a consequence, the nature of the substrate does not matter any more. The design problem therefore consists to find a broadband antireflection coating for a metallic substrate. This metallic substrate acts of course as a mirror and

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we need to force light to penetrate this mirror. The solution is to place an equivalent mirror in front of the metallic layer in order to form a Fabry-Perot structure. After the deposition of a dielectric spacer layer, we can deposit as we like a dielectric or a metallic mirror. Instead of the transmission peak we can observe with a classical Fabry-Perot filter, we obtain in that case an absorption peak. As usually, the spectral width of this absorption peak is directly related to the mirror reflectance, as illustrated in figure 2. As a consequence, low reflecting metals, such as chromium or nickel, are preferable to obtain broadband absorption properties. A second conclusion illustrated in figure 2 is that the use of a second metallic mirror leads to more achromatic absorption properties than the use a dielectric mirror. Notice that in that case the absorption is not only located inside the metallic substrate but distributed in both metallic layers. This classical approach permits to understand the behavior of the component and to justify the choice of the metallic material. Such simple design structures can be used to manufacture solar cells for example [5].

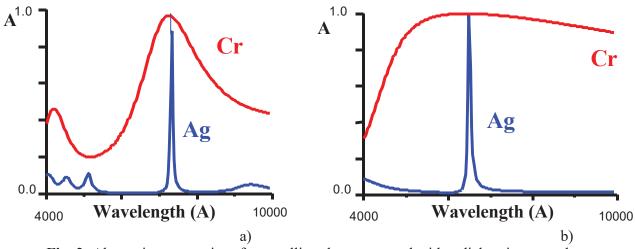


Fig. 2: Absorption properties of a metallic substrate coated with a dielectric spacer layer, followed by a dielectric mirror (a) or a metallic mirror (b) in the case of chromium (Cr) or silver (Ag). Metal-dielectric structures, using low reflecting metals, give more achromatic result.

However, the high absorption spectral range is still too short for our need. In order to obtain a broadband component, we need to increase the layer count, still using alternated dielectric and metallic layers. At this point, numerical optimization permits to determine easily the layers thickness to obtain maximum efficiency. The achromatic behavior of such a coating is much more difficult to investigate and will not be discussed here [6]. However, it may be useful to compare these coatings with classical broadband antireflection coatings. Such antireflection coatings can be easily designed for high refractive index substrates such as ZnSe or Ge for infrared applications. The main idea is to design a coating that acts as a graded index layer. The layer refractive index corresponds to the substrate at the beginning of the deposition and to the external medium index at the end. Such a layer can be manufactured using the co-evaporation of two materials. However, using classical design techniques, such as equivalent symmetrical stack [7], this graded index profile can be exchanged with a more common multi-layer index profile using two materials. In both cases, we need high and low index materials that are respectively close to the substrate and the external medium to reach the highest efficiency.

Broadband metal-dielectric absorbers can be considered similarly. Indeed, such a coating acts as an anti-reflection coating deposited on a "high index" metallic substrate. The "high index" material for the coating is the metal itself. As a consequence, we can intuitively expect better results using a low index dielectric material to mach adequately the external medium. To illustrate this statement, figure 3 gives the optimum result for a four layer design using chromium either with a high index

dielectric material such as zinc sulfide (n = 2.3) or with a low index dielectric material such as cryolite (n = 1.3).

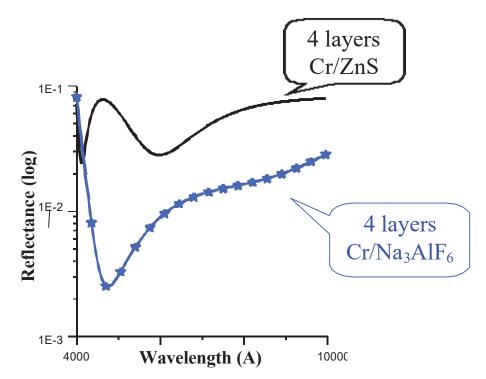


Fig. 3: Reflectance of 4-layer designs using chromium and either zinc sulfide or cryolite. Low index dielectrics lead to lower reflectance levels, which corresponds to maximum absorptance since transmittance is zero.

Figure 4 shows that an increase of the layer count tends to improve the efficiency. Indeed, a six layer chromium / cryolite design permits to reach 99.9 absorptance over the visible range.

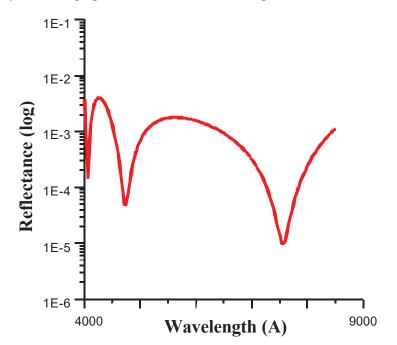


Fig. 4: Reflectance of 6-layer design using chromium and cryolite.

However, the layer count is not the only parameter that determines the coating efficiency. Higher absorption levels require higher precision in the layer thickness determination, which becomes unrealistic considering the thickness monitoring accuracy during manufacturing. Figure 5 shows the influence of thickness errors on the 6-layer design. As one can see, reflectance may increase rapidly, which is typical of antireflection coatings.

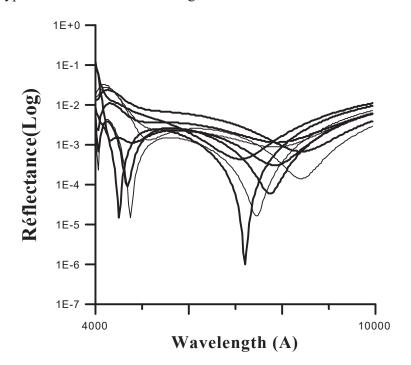


Fig. 5: Evolution of the reflectance of the design given in fig.4 considering random errors on layer thicknesses (10% max. relative error for each layer).

To conclude this section, a low reflecting metal associated with a low index dielectric material permit to design efficient broad-band coatings using less than ten layers. However, the metal refractive index used in this section was taken in the literature and concerns bulk material [8]. To obtain optimum efficiency, we need to determine the metal index in thin film form, for our deposition conditions. Moreover, notice that the first metallic layer of the design (given in figure 5) is opaque while the last one is only 3 nanometers thick, which implies to study the refractive index of metallic layers for both opaque and semi-transparent films. This is the subject of the next section.

3- INDEX DETERMINATION FOR METALLIC FILMS.

The index determination of metallic films can be achieved using several techniques such as ellipsometry or surface plasmon technique [9]. However, in order to avoid the development of new measurement apparatus, we preferred to develop a new index determination method based on classical reflectance and transmittance spectrophotometric measurements. We will consider first the case of opaque metallic layers and then semi-transparent layers.

3-1) Opaque metallic layer.

We only consider in this subsection the particular case of opaque layers. As a consequence, the thickness of the layer does not matter. We only need to determine the complex refractive index $(n_m-i.k_m)$ of the film. In order to determine these to quantities, we need at least two independent measurements. The first method we used simply consists to measure the reflectance of the sample on both sides, namely in the air and inside the substrate that is assumed to be absorption free. From

these two quantities, one can extract easily the complex refractive index of the film. However, this method is sensitive to noise and to the substrate/film interface quality. In particular, the results we obtained in the case of chromium were very far from the literature and we could observe a slight migration of chromium inside the substrate by electron microscopy. Because of this anomaly with chromium, we preferred to go on using nickel.

More over, in order to avoid interface problem, we developed a new method which requires two measurements in the air side. The reflectance of the metallic layer is measured first. Then, a thick dielectric layer is deposited on the metallic film and the second measurement corresponds to the reflectance of this new sample, as illustrated in figure 6.

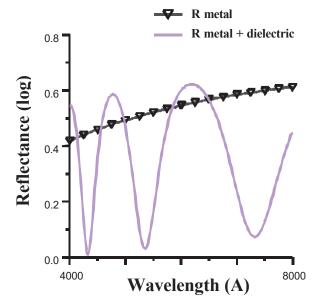


Fig. 6: Measured reflectance of an opaque nickel layer before and after the deposition of a thick dielectric layer.

The dielectric layer is also deposited on a bare substrate in order to determine its refractive index and thickness. At last, a numerical optimization procedure determines the refractive index of the metallic film that explains the measured quantities. We successfully applied this method with chromium and nickel. The results we obtained with chromium were much closer from the literature than those we obtained with the first method mentioned above. More over, this new method is far less sensitive to measurement noise. The complex refractive index we obtained with nickel is given in figure 7.

3-2) Semi-transparent metallic layers.

In the case of semi-transparent metallic layers, we need to determine the complex refractive index of the metallic film as well as its thickness. We therefore require at least three independent measurements. As previously we can use reflectance measurements in the air, on the metallic film first and after deposition of a dielectric layer. However, since the sample is now semi-transparent, we can also use transmittance measurements, before and after the dielectric layer deposition. Using these four quantities, we can use as previously a numerical optimization procedure to determines the characteristics of the metallic layer. This method was applied with three nickel layers with transmittance levels of 70, 50 and 20 percents. The results we obtained are given in figure 8. The main conclusion is that the refractive index strongly depends on the layer thickness. This may be explained by the evolution of the film packing density at the beginning of the deposition. However, all the calculations we made assumed the films to be homogeneous and one conclusion is that this

model is not adequate for this study. As a consequence, the index values we found are approximate results. In order to overcome this contradiction, we developed a graded index layer model.

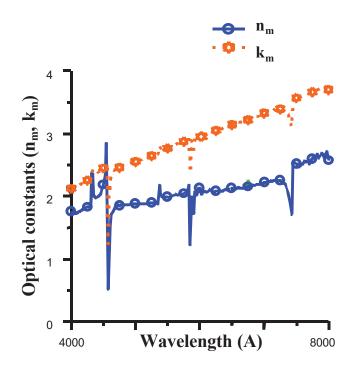


Fig. 7: Refractive index and extinction coefficient of opaque nickel . (Determined from measurements given in figure 6)

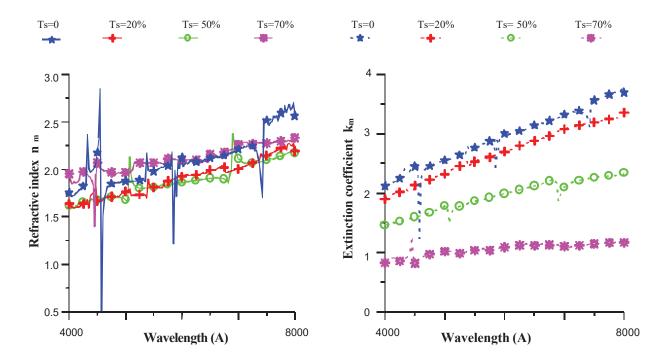


Fig. 8: Refractive index and extinction coefficients determined for opaque and semi-transparent nickel layers (Transmittance levels of 70,50,20 and 0 %).

3-3) Graded index metallic layer.

In order to take account of the film index evolution versus thickness, we consider that the film packing density increases during deposition, from zero to unity. The film is therefore considered as a mixture of metal and air and its index is calculated using Maxwell Garnet theory [10]. According to this theory, figure 9a gives the evolution of the complex refractive index of a nickel/air mixture versus the packing density at wavelength 600 nm. For more simplicity, we assumed that the maximum packing density is reached for an opaque layer so that the index we determined in that particular case corresponds to the metal index. For the packing density evolution, we propose the following model:

$$Q = \frac{2}{\pi} \arctan\left(\frac{d}{\alpha}\right)$$

where Q is the packing density and d the film thickness. The α coefficient must be chosen adequately in order to explain as well as possible the reflectance and transmittance optical properties measured for semi-transparent layers. In our case, a correct value is $\alpha = 0.973$ nm.

Figure 9b gives the evolution of the packing density versus thickness. The thicknesses corresponding to the semi-transparent nickel layers we studied are indicated on this figure.

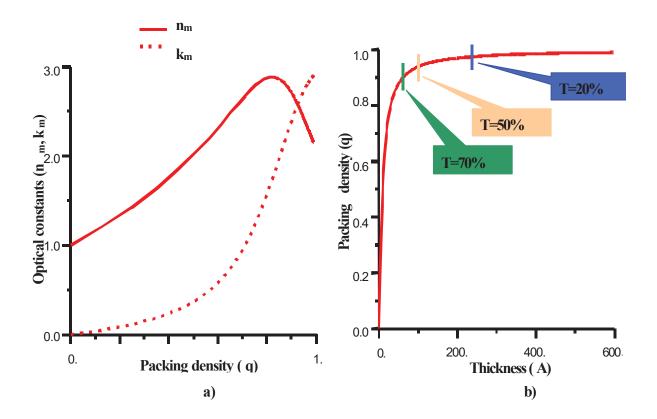


Fig. 9: Evolution of the optical constants of a nickel layer versus the film packing density at wavelength 600 nm (a) and evolution of the film packing density versus the film thickness (b)

4- MANUFACTURING OF HIGH ABSORPTION METAL DIELECTRIC ABSORBERS

Using the design principles given in section 2, using the graded index metallic layer model developed in section 3, we designed a broad band nickel-cryolite absorber for the visible range. The thicknesses (expressed in nm) of the layers are the following:

Substrate / Ni (200) Cryolite (64.8) Ni (20.3) Cryolite (72.7) Ni (10.4) Cryolite (89.4) / air

The deposition technique we used is classical electron beam evaporation. Quartz crystal monitoring was used to control the deposited thicknesses. However, the calibration of the quartz crystal monitor was performed with optical monitoring during several preliminary depositions.

Figure 10 gives the calculated reflectance of the design and the measured reflectance of two coatings. Obviously, the thickness monitoring is the critical point to obtain optimum results.

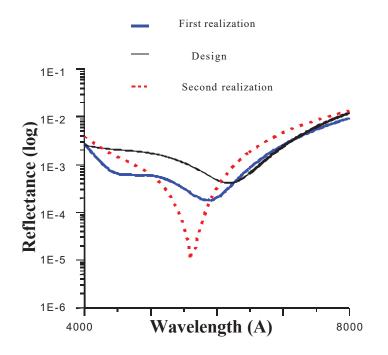


Fig. 10: Calculated and measured reflectance of a 6 layer Ni/Cryolite broadband absorber: Absorption is measured higher than 0.999 over the major part of the visible range.

5- CONCLUSION

We described in this paper the design and manufacture of high absorption metal dielectric coatings. First, we give the design principle to obtain broadband absorption properties. The main result is that we must combine a low reflecting metal with a low index dielectric. As in the case of antireflection coatings, the efficiency is linked to an accurate knowledge of the thin film material indices. To fulfill this requirement, we developed new methods to determine the complex refractive indices of metallic materials in thin film form. These methods were applied with opaque and semi-transparent nickel layers. The results showed a great dependence of the film index versus its thickness, probably due to the evolution of the film packing density during deposition. As a consequence, we developed a graded index model to take account of this phenomenon. Using this model, we designed a coating for the visible range. The manufacture showed that the thickness monitoring was a critical point to obtain high efficiency. At present time, the best coating we manufactured provides absorption higher than 99.9 % from 400 to 650 nm.

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