Symmetrical periods in antireflective coatings for plastic optics

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Plastic optical parts require antireflective as well as hard coatings. A novel design concept for coating plastics combines both functions. Symmetrical three-layer periods with a phase thickness of $3/2\pi$ are arranged in a multilayer to achieve a step-down refractive-index profile. It is shown mathematically that the equivalent index of symmetrical periods can be lower than the lowest refractive index of a material used in the design, if the phase thickness of the symmetrical period is set equal to $3/2\pi$ instead of the usual $\pi/2$. The straightforward application of the concept to the design of antireflection coatings in general is demonstrated by example. © 2003 Optical Society of America

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1. Introduction

Hard coatings with a physical thickness of at least 1 μm are required for plastic optics and plastic display windows to make them scratch resistant. Frequently, an antireflection (AR) function is required in addition. Abrasion-resistant AR coatings are well known in the field of ophthalmic lenses.[1] Most of the hard coatings used in lenses are lacquers based on siloxane. Usually, a classical four-layer AR coating is deposited by a physical vapor deposition process on top of a single hard coating.[2] Our efforts were aimed at developing an AR coating for plastic substrates that would be scratch resistant. As a result of our design investigations, a special type of AR design with a quasi-periodic structure was obtained.[3] The AR-hard design type is characterized by thin high-index layers that are more or less evenly spaced by thick low-index layers. The calculated spectral performances and index profiles of some AR-hard designs are listed in Fig. 1. Coating results obtained by plasma ion-assisted deposition on polymer substrates are described elsewhere.[4] Our aim here is to discuss the quasi-periodic AR-hard design within the context of an equivalent-layer concept. In Section 2 we analyze the AR-hard design type with a view to the basics of symmetrical periods and equivalent layers. In Section 3 we derive an algorithm to obtain the AR-hard design and apply an example.

2. Design Analysis

Based on the condition to realize a thick hard coating with an inherent AR function for plastic substrates, we began all our design approaches with a single low-index layer of 1-μm thickness. The target value for residual reflection in the visible range was set to approximately 0.3% or, in any case, greater than zero.

To create suitable AR designs we applied the needle optimization technique, which permits a target to be approached in steps by adding new layers to a given design.[5] A so-called P function is calculated to indicate where, within the design, the incorporation of an additional layer best improves the design performance. Typically, the additional layers (known as needle layers) are thin. In this way, designs of the AR-hard type with total physical thicknesses from approximately 1 μm to greater than 3 μm have been achieved. Examples are shown in Fig. 1.

For a discussion of the properties of the design type, let the typical structure of the AR-hard type be represented by a nine-layer system known as AR-hard-9:

$$\begin{aligned}&\text{sub}/2.433L\ 0.144H\ 2.83L\ 0.226H\\&\qquad 2.704L\ 0.366H\ 2.55L\ 0.534H\ 1.233L/\text{air},\end{aligned}$$

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where L and H are low-index and high-index layers with $n_L = 1.46$ and $n_H = 2.1$, respectively. Optical thicknesses are given in fractions of one quarter wave at the reference wavelength λ_0. Sub stands for the substrate and air for the radiant incidence medium, with $n_{\text{sub}} = 1.49$ and $n_{\text{air}} = 1$. For simplification, normal incidence and the absence of any losses is assumed. At the wavelength centroid of $\lambda_0 = 516$ nm, the physical thicknesses in terms of nanometers of the AR-hard-9 design are

sub/215.8L 7H 250L 13.9H 239L 22.5H 225.3L 32.8H 108.9L/air.

The total thickness of the multilayer stack is 1115.2 nm, and the total thicknesses of the low-index and the high-index layers are 1039.1 and 76.2 nm, respectively. The coating reduces the residual reflectance to an average value of 0.23% in the spectral range from 430 to 645 nm.

The typical quasi-periodic structure and the optical performance of AR hard can be explained if we apply the concept of symmetrical periods.[6–8] Any combination of thin films that is symmetrical (i.e., one in which the sequence of layers is unchanged when they are listed in reverse order) can be represented mathematically by a single equivalent film having an equivalent index N and an equivalent phase thickness Γ. Both are available from indices n_1 and n_2 and phase thicknesses ϕ_1 and ϕ_2 of the individual layers. The equivalent refractive index N of a symmetrical nonabsorbing three-layer period is given by

$$N^2 = {n_1}^2 \left[\frac{\sin 2\phi_1 \cos \phi_2 + \frac{1}{2}\left(\frac{n_1}{n_2} + \frac{n_2}{n_1}\right)\cos 2\phi_1 \sin \phi_2 - \frac{1}{2}\left(\frac{n_1}{n_2} - \frac{n_2}{n_1}\right)\sin \phi_2}{\sin 2\phi_1 \cos \phi_2 + \frac{1}{2}\left(\frac{n_1}{n_2} + \frac{n_2}{n_1}\right)\cos 2\phi_1 \sin \phi_2 + \frac{1}{2}\left(\frac{n_1}{n_2} - \frac{n_2}{n_1}\right)\sin \phi_2} \right]. \tag{1}$$

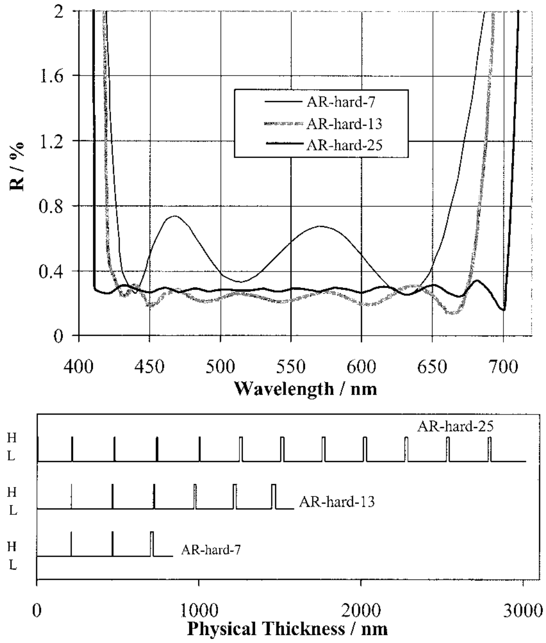

Fig. 1. Index profiles and performance of AR-hard coatings consisting of 7 (AR-hard-7), 13 A(R-hard-13), and 25 (AR-hard-25) layers.

The equivalent phase thickness Γ is defined in terms of its cosine

$$\cos \Gamma = \cos 2\phi_1 \cos \phi_2 - \frac{1}{2}\left(\frac{n_1}{n_2} + \frac{n_2}{n_1}\right)\sin 2\phi_1 \sin \phi_2, \tag{2}$$

where

$$\phi_1 = \frac{2\pi}{\lambda} n_1 d_1, \tag{3a}$$

$$\phi_2 = \frac{2\pi}{\lambda} n_2 d_2, \tag{3b}$$

with physical layer thicknesses d_1 and d_2, respectively. In most practical cases, Γ is simply proportional to the period thickness $(2\phi_1 + \phi_2)$, with the proportionality constant being close to unity.[9] It should be noted that the concept of equivalent layers describes a mathematical equivalence and not a physical one; i.e., both N and Γ change with wavelength but in a mathematical sense only.

The AR-hard-9 design can be rearranged by splitting each of the thick low-index layers into two parts of different thicknesses, except for the layer next to air. In this way, four symmetrical periods are obtained (Table 1):

sub/1L(1.433L 0.144H 1.433L)

×(1.387L0.226H 1.387L)(1.317L 0.366H 1.317L)

×(1.233L 0.534H 1.233L)/air.

We calculated equivalent indices and equivalent phase thicknesses for each period by using Eq. (1) with refractive indices n_L and n_H for n_1 and n_2, respectively. The thicknesses of the periods are shown in Table 1. Each original period equals three quarter-wave optical thicknesses (3-QWOTs), and all equivalent phase thicknesses calculated equal nearly $3/2\pi$. Starting with the substrate, the first low-index layer L and the following equivalent layers

Table 1. AR-hard-9 Design[a]

Layer	Material	Design AR-hard-9 (QWOT)	AR-hard-9 Rearranged (QWOT)	Material or Equivalent Layer	Optical Thickness (QWOT)	Equivalent Index N' at 516 nm	Equivalent Phase Thickness (Units of $\pi/2$)
1	L	2.443	1.000	L	1		1
			1.443				
2	H	0.114	0.114	A'	3.00	1.3666	3.118
3	L	2.830	1.443				
			1.387				
4	H	0.226	0.226	B'	3.00	1.2835	3.081
5	L	2.704	1.387				
			1.317				
6	H	0.366	0.366	C'	3.00	1.1944	3.098
7	L	2.550	1.317				
			1.233				
8	H	0.534	0.534	D'	3.00	1.1111	3.075
9	L	1.233	1.233				

[a] From substrate site to air and split into its component periods and optical properties of equivalent layers A', B', C', and D'.

build up a layer stack with decreasing refractive indices matching the refractive index of the substrate to that of air. Thus, the AR-hard-9 design, obtained originally by an optimization procedure, represents a typical step-down design: sub/1L $3A'$ $3B'$ $3C'$ $3D'$/air. Figure 2 shows the equivalent indices of A', B', C', and D' versus wavelength calculated at $\lambda_0 = 516$ nm. Note that this dispersion results from the dependency of N on the phase thickness of the period that changes with wavelength according to Eq. (1). Figure 3 shows the optical performance of the step-down design compared with the AR-hard-9 in its original form.

The use of equivalent layers in AR coatings is a well-known design method. In 1952, the first example was given by Epstein in his fundamental paper about the design of optical filters.[8] In 1962, Berning suggested the use of symmetrical periods for AR purposes.[9] Nevertheless, Berning focused on AR coatings on high-index infrared optical materials. His periods represent equivalent indices within the range $n_1 < N < n_2$, and it does not seem possible to substitute a layer with an equivalent index lower than n_1 by a symmetrical period. In addition, there are other step-down AR coatings for high-index substrates described in the literature for which the restriction $n_1 < N < n_2$ is valid.[10,11]

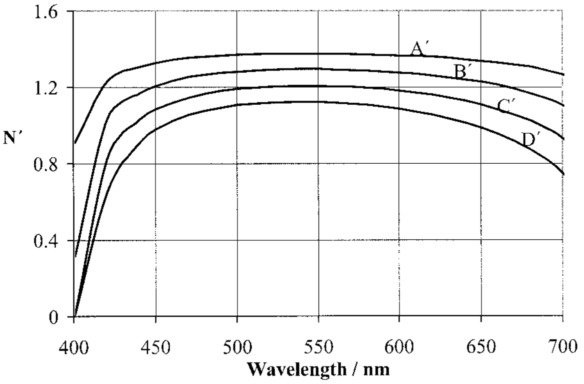

Fig. 2. Dispersion of the equivalent layers A', B', C', and D' belonging to the symmetric periods of the AR-hard-9 design for a design wavelength of 516 nm.

Furthermore, substitution for QWOT layers of an unobtainable refractive index has been a commonly applied design approach since Ohmer published the equations

$$\sin \phi_2 = \left(\frac{n_1/N - N/n_1}{n_1/n_2 + n_2/n_1}\right)\sin \Gamma, \tag{4}$$

$$\cot 2\phi_1 = \frac{1}{2}\left(\frac{n_1}{n_2} + \frac{n_2}{n_1}\right)\tan \phi_2 \tag{5}$$

for the straightforward calculation of the phase thicknesses necessary to build up symmetrical periods.[12] Given the refractive indices n_1 and n_2 and with the equivalent phase thicknesses set to an odd multiple of $\pi/2$, the phase thicknesses ϕ_1 and ϕ_2 of the individual layers can be calculated to synthesize a de-

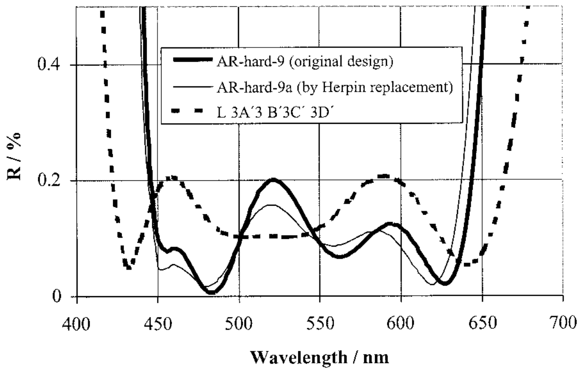

Fig. 3. Reflectance versus wavelength of the AR-hard-9 design: sub/2.433L 0.144H 2.83L 0.226H 2.704L 0.366H 2.55L 0.534H 1.233L/air and for the corresponding step-down design: sub/1L $3A'$ $3B'$ $3C'$ $3D'$/air (the dispersion of equivalent layers shown in Fig. 2 is considered). Design AR-hard-9a: sub/2.443L 0.106H 2.823L 0.226H 2.687L 0.366H 2.529L 0.534H 1.222L/air was achieved after Herpin replacement of equivalent layers A', B', C', and D'.

sired equivalent index N. The $\sin \Gamma$ in Eq. (4) can achieve a value of $+1$ or -1. Formulas for what is called the Herpin index have been implemented in thin-film software, for example, the Essential Macleod.[13] However, in all the current commercially available software, the condition $\sin \Gamma = +1$ is used for the application of Eq. (4) only. Therefore, the synthesis of an equivalent refractive index lower than the given n_1 is not yet possible by means of design software.

The period thickness is given by

$$2\phi_1' + \phi_2 = 3\pi/2 \text{ (or uneven numbers of } 3\pi/2). \tag{8}$$

Equation (1) can be simplified for the 3-QWOT period by use of Eq. (7) and the trigonometric relations $\sin 2(\phi_1 + \pi/2) = -\sin 2\phi_1$ and $\cos 2(\phi_1 + \pi/2) = -\cos 2\phi_1$. The square of the effective index N' for the 3-QWOT period is then given by

$$N'^2 = n_1^2 \left[\frac{-\sin 2\phi_1 \cos \phi_2 - \frac{1}{2}\left(\frac{n_1}{n_2} + \frac{n_2}{n_1}\right)\cos 2\phi_1 \sin \phi_2 - \frac{1}{2}\left(\frac{n_1}{n_2} - \frac{n_2}{n_1}\right)\sin \phi_2}{-\sin 2\phi_1 \cos \phi_2 - \frac{1}{2}\left(\frac{n_1}{n_2} + \frac{n_2}{n_1}\right)\cos 2\phi_1 \sin \phi_2 + \frac{1}{2}\left(\frac{n_1}{n_2} - \frac{n_2}{n_1}\right)\sin \phi_2} \right]. \tag{9}$$

3. Transformation Formula

Figure 4 shows the periodic variations of the equivalent index versus phase thickness of the three-layer period D' if only the phase thickness ϕ_1 of the outer layers is increased whereas that of the inner layer is held constant. Only real values of N around a period thickness of $\pi/2$ and multiples of $\pi/2$ are shown. To simplify the work with equivalent layers it would be helpful to look at the solutions of Eq. (1) for both cases, i.e., phase thicknesses of $1/2\pi$ (QWOT period) and $3/2\pi$ (3-QWOT period). For the phase thickness of the QWOT period we get

$$2\phi_1 + \phi_2 = \pi/2. \tag{6}$$

The behavior of the 3-QWOT period can be described mathematically by adding the value of $\pi/2$ to the phase thickness ϕ_1:

$$\phi_1' = \phi_1 + \pi/2. \tag{7}$$

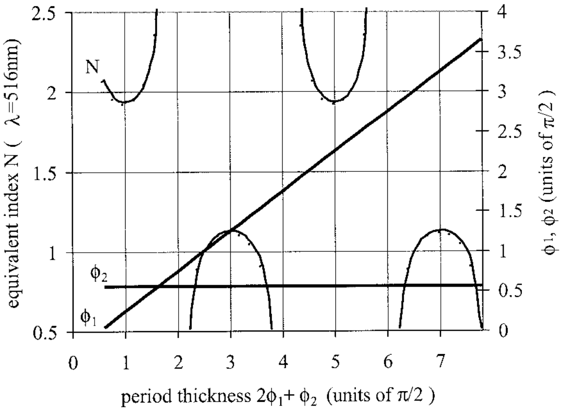

Fig. 4. Equivalent index N of a symmetric period ϕ_1L ϕ_2H ϕ_1L depending on period thickness at wavelength 516 nm (example: equivalent layer D' with $\phi_2 = 0.534 = \text{const.}$).

Comparison of Eqs. (1) and (9), yields the dependency of N' on N:

$$N' = n_1^2/N. \tag{10}$$

Equation (10) yields the equivalent index N' of a symmetric 3-QWOT period if N is the equivalent index of the QWOT period for the phase thickness conditions defined in Eqs. (6)–(8). The phase thickness of the period changes from $\pi/2$ to $3/2\pi$, whereas the thickness of the middle layer of the period does not change. In the case discussed here ($n_1 < n_2$), there is a restriction for N' because N maximally equals n_2:

$$N' \min = n_1^2/n_2 \quad \text{if} \quad n_1 < n_2. \tag{11}$$

Below, a fundamental way is shown for the straightforward calculation of periods having indices N' lower than n_1. A desired index N' can readily be transformed to the Herpin index N. The phase factors for a symmetric LHL period are then available from design software. In the next step, the period achieved has to be enlarged by adding quarter-wave L layers before and after the QWOT period.

As an example, let the design technique described be applied to obtain designs such as AR hard without optimization techniques. With Eq. (10) we were able to rearrange the AR-hard-9 design to

sub/2L 1A 2L 1B 2L 1C 2L 1D 1L/air,

with the corresponding values in Table 2. First, the equivalent indices N of the QWOT periods corresponding to the periods A', B', C', and D' (Table 1) can be calculated by application of Eq. (10). Now, thickness values for L and H layers are available by common Herpin replacement of the equivalent indices N by use of software.[9] After adding quarter-wave L layers to both L layers of each period, the final AR-hard-9a design is achieved. An identical result can be obtained by use of Eqs. (4) and (5) directly, but with the condition of $\sin \Gamma = -1$.

Performance of the AR-hard-9a design is shown in Fig. 3. There is a small difference between the val-

Table 2. Rearranged AR-hard-9 Design[a]

Layer	Material or Equivalent Layer	Equivalent Phase Thickness (Units of $\pi/2$)	Equivalent Index N at 516 nm	Material	Optical Thickness by Herpin Replacement (QWOT)	Material	AR-hard-9a Obtained by Herpin Replacement (QWOT)
1	L	1.000		L	2	L	2.439
				L	0.439		
2	*A*	1.000	1.5598	H	0.114	H	0.114
				L	0.439		
3	L	2.000		L	2	L	2.819
				L	0.380		
4	*B*	1.000	1.6607	H	0.226	H	0.226
				L	0.380		
5	L	2.000		L	2	L	2.687
				L	0.307		
6	*C*	1.000	1.7846	H	0.366	H	0.366
				L	0.307		
7	L	2.000		L	2	L	2.529
				L	0.222		
8	*D*	1.000	1.9185	H	0.534	H	0.534
				L	0.222		
9	L	1.000	1.46	L	1	L	1.222

[a] Equivalent indices N of its QWOT periods A, B, C, and D. The AR-hard-9a design is achieved after Herpin replacement of A, B, C, and D.

ues of the original AR-hard-9 and the synthesized values that results from the difference between the equivalent phase value of approximately $3/2\pi$ given by the analysis and the exact $3/2\pi$ value that is used for the synthesis.

As another example to demonstrate the design principle, let us regard a step-down AR coating implemented by use of a so-called maximally flat formula from Thelen.[14] This formula is an algorithm to calculate the indices for layers of identical optical thickness for building up an optimal step-down AR coating. We chose a five-layer sequence sub/L 3F′ 3G′ 3J′ 3K′/air similar to the AR-hard-9. The index profile of this step-down design compared with the AR-hard-9 design is shown in Fig. 5. It should be pointed out that an equivalent phase thickness of $3\pi/2$ is necessary for layers F′, G′, J′, and K′. Otherwise, an equivalent index lower than n_L is not available. The values for N and the optical thicknesses for L and H layers calculated by use of Eqs. (4), (5), and (10) are shown in Table 3. Figure 6 shows the performances of the maximally flat design by use of constant refractive indices and after replacement of layers $3F' \; 3G' \; 3J' \; 3K'$ by symmetrical periods LHL.

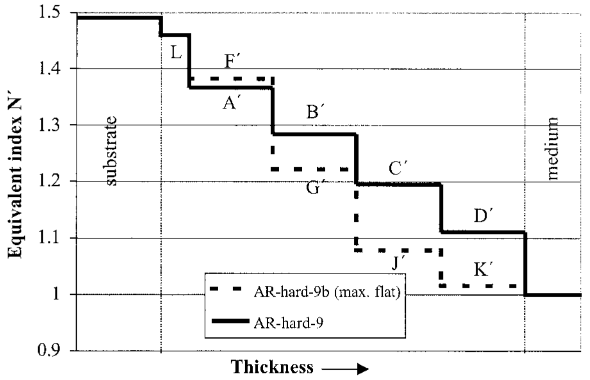

Fig. 5. Index profiles of step-down AR coatings: layer sequence L *A′ B′ C′ D′* corresponding to the equivalent layers that build up design AR-hard-9 and layer sequence L *F′ G′ K′ J′* with refractive indices calculated by use of a so-called maximally flat formula.[12]

4. Conclusion

Antireflection coatings of the AR-hard type can be understood as an arrangement of symmetrical three-layer LHL periods. Each period can be interpreted as an equivalent layer with three QWOTs. The equivalent layers build up a layer stack that matches the refractive index of the substrate with that of air. It has been demonstrated that symmetrical LHL periods of three QWOTs can be applied to replace layers with unobtainable refractive indices lower than n_L. The mathematical relation between a QWOT period and the same period enlarged by adding quarter-wave L layers as outer layers has been derived. It is evident that similar considerations are possible for periods of HLH structure to achieve equivalent refractive indices higher than n_H.

However, the total physical thickness of coatings obtained by this design technique is high compared with that of other designs that can give a comparable performance. Thinner coatings are more practical for many applications and mostly available by other design techniques. A practical advantage of the design type described here is obvious if the coating has to be thick, for example, on plastic components, to make them scratch resistant. For coatings for which a total thickness of 2 μm or more is desired, the antireflective performance can be broadened compared with the AR-hard-9 design. In addition, the low volume of high-

Table 3. Step-Down Design sub/L 3F′ 3G′ 3J′ 3K′/air[a]

Material or Equivalent Layer	Equivalent Phase Thickness (Units of $\pi/2$)	Equivalent Index N'	Equivalent Index N	Material	Optical Thickness by Herpin Replacement (QWOT)	Material	AR-hard-9b Max. Flat Design (QWOT)
L	1	1.46		L	1.000	L	2.450
F′	3.00	1.3830		L	1.000		
			1.5413	L	0.450		
				H	0.093	H	0.093
				L	0.450	L	2.780
				L	1.000		
G′	3.00	1.2210		L	1.000		
			1.7458	L	0.330		
				H	0.321	H	0.321
				L	0.330	L	2.509
				L	1.000		
J′	3.00	1.0780		L	1.000		
			1.9774	L	0.179		
				H	0.622	H	0.622
				L	0.179	L	2.179
				L	1.000		
K′	3.00	1.0151		L	1.000		
			2.1000	L	0.000		
				H	1.000	H	1.000
				L	0.000	L	1.000
				L	1.000		

[a] Design consists of layers with 3-QWOT and unobtainable refractive indices. The equivalent indices N' of the 3-QWOT periods and the corresponding indices N (after removing the outer quarter-wave L layers from each period) are shown in columns 3 and 4. Design AR-hard-9b (columns 7 and 8) is achieved by Herpin replacement.

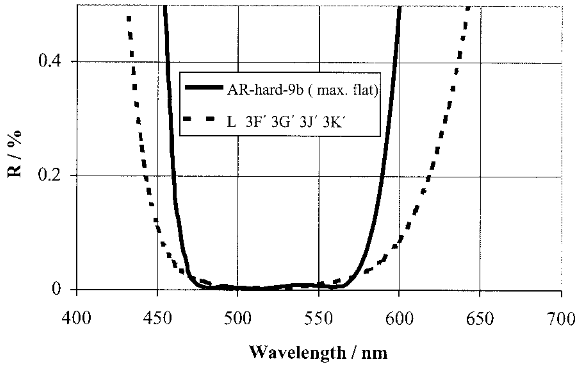

Fig. 6. Reflectance versus wavelength of step-down design sub/1L 3F′ 3G′ 3J′ 3K′/air and of the corresponding design AR-hard-9b after Herpin replacement: sub/2.450L 0.093H 2.780L 0.321H 2.509L 0.622H 2.179L 1H 1L/air.

index material in coatings of the AR-hard type helps to make the deposition process for heat-sensitive materials such as polymers as cold as possible, because of the high thermal output of high-index materials during evaporation.

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