THE IMPACT OF COMPUTERS ON THE DESIGN AND MANUFACTURE OF OPTICAL MULTILAYER COATINGS DURING THE PAST 50 YEARS

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ABSTRACT
Fifty years ago, with very few exceptions, optical multilayer coating designs were composed of layers of quarter wave optical thicknesses. To obtain a given performance, thin film designers usually varied the number of layers and the refractive indices in a multilayer. Design methods based on vectors, and on various charts and diagrams, were often too tedious to apply when problems needed to be specified at many wavelengths and were at the same time complex enough to require a larger number of layers for their solution. Also, the monitoring of the thicknesses of non-quarter wave layers was difficult then. Philip Baumeister’s 1958 paper in which he described the use of a computer optimization method to refine the performance of an approximate starting design to obtain a short wavelength cut-off filter was a major breakthrough in our field. However, the solution asked for the use of non-quarter wave layers. This work was followed by further developments in numerical design as well as in more sophisticated monitoring procedures, also based on the use of computers. Since that time, progress in numerical methods, deposition techniques and in-situ measurement has made possible the reliable automatic manufacture of optical thin film systems with very complex spectral transmittance or reflectance characteristics consisting of many tens, or even hundreds of layers. In this talk an attempt will be made to review these developments.

DESIGN AND MANUFACTURE OF OPTICAL MULTILAYERS IN 1957
Before 1957 most optical multilayer systems consisted of quarter wave- or multiple quarter wave layers. The most important exceptions were two-layer V-type antireflection coatings and metal-dielectric Fabry-Perot interference band pass filters. There were two impediments to the use of non-quarter wave systems: the design and the monitoring of the thicknesses of such layers.

The two-by-two matrix representation of a layer for the calculation of the properties of thin film systems was well-known by then so, theoretically, calculations on non-quarter wave systems could have been performed [1,2]. However, computers were not widely available then and performing calculations on anything but the simplest systems was tedious with hand-cranked or electrical calculators. The tools available for the design of optical thin film systems at the time included Smith and Kard charts, admittance and circle diagrams, and vector methods [3,4]. These methods were very useful for systems consisting of a few layers, and they provided a lot of insight into the way such systems worked, but they were sometimes very approximate. Exact explicit expressions for the reflectances of one, two and three-layer systems were published [5-7]. For systems consisting of more layers, various series expansions for systems of equal optical thickness which yielded the refractive indices of the individual layers have been described, as have expressions for the reflectances of periodic systems [3,4]. But these methods were still too cumbersome to use on problems in which transmittances \( T \) or reflectances \( R \) were specified at very many different wavelengths and when the systems consisted of many layers. Furthermore, they often asked for refractive indices that did not exist in nature. The powerful Herpin equivalent index concept was first described in 1947 [8, 9]. Nevertheless, with the above tools most of the generic filter types that we know today had been designed and produced. However, the systems were two-material quarter- or multiple quarter wave solutions, except for antireflection coatings in which more than two materials were used.

In 1957 optical thin film systems were mostly deposited by evaporation, a process that in those days was not very well controlled. This necessitated the use of in-situ measurements of the layer thicknesses. Quartz crystal monitors at the time were more primitive than they are today. They did not provide a direct measurement and so required calibration and, like all indirect methods, they were not very reliable in the presence of fluctuations in the evaporant plume. The direct optical monitoring of layers of quarter wave thickness was simpler because the end-point of the deposition of each layer occurred at a maximum or minimum of \( T \) or \( R \) (Figure 1a). Thus this method did not require any absolute measurements. Even though at the termination point the measurement was least sensitive to thickness errors, Macleod and Pelletier showed later that a thickness error compensation mechanism occurs that leads to good results [10]. This effect it is still of great importance to-day. Certain narrow-band high rejection telecom filters consist of in excess of a hundred layers and they can only be successfully produced if the thicknesses of certain groups of layers in such systems are controlled by the quarter wave monitoring method.

Figure 1. Control of layer thicknesses, a) quarter wave monitoring, b) most sensitive wavelength method [34]. continued on page 36
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PROGRESS IN OPTICAL THIN FILM DESIGN
The use of only quarter wave layers in the design of non-absorbing optical multilayer coatings came at a great cost. The performance of a multilayer depends on its construction parameters,

\[ n_m, n_s, d_i, n_i, i=1, 2, \ldots L \]

and on the overall thickness of the multilayer. Here \( n_m, n_s \) are the refractive indices of the incident medium and substrate, and \( d_i, n_i \) are the metric thickness and refractive index of the \( i \)-th layer of an \( L \)-layer system. When one chooses to deposit only quarter wave layers, one thus neglects half of the available construction parameters. This is serious, because the quality of a solution depends on the overall thickness of the layer system and on the number of construction parameters.

With regard to the status of refractive indices as variables in thin film design, Alexander Tikhonravov has shown in 1987 with his maximum principle that, for problems with normal incidence of light and for solutions having the same optical thickness, the performance is not improved if more than the two extreme refractive indices of the available coating materials are used [11,12]. However, in the two-material solutions, the total number of layers will usually be larger and some of the individual layer thicknesses may be smaller. This follows intuitively from the Herpin equivalent index concept [8, 9]. The maximum principle does no longer hold if the problem is specified for non-normal incidence of light polarized in more than one direction.

Refinement
When computers became more widely available, they were first used for the analysis of the spectral and angular properties of more complex layer systems. Some people started to perform trial-and-error calculations in which the thicknesses of some layers of the system were varied in a systematic manner. But the real breakthrough came in 1958 when Philip Baumeister showed how, by using computer refinement methods, the secondary reflectance ripples of a quarter wave stack could be dramatically reduced to produce a good cut-off filter without the need to increase the number of layers (Figure 2) [13]. Baumeister used the damped least squares method in his optimization.

Figure 2. Baumeister’s cut-off filter produced by the refinement of layer thicknesses. Calculated performances (a), (b) and the construction parameters (Table 1) before and after refinement [13].

My system doesn’t need to work at any cost...

My system needs to work with all costs in mind.
Since that time, many more optimization methods have been used for optical thin film design. These can be classified into methods in which zero, first and second order derivatives are evaluated during the refinement process. Most refinement methods require the minimization of a single-valued function that represents at any moment of the design process the performance of the system. Many people call this function the merit function \( M \), others prefer the term error function. Quite frequently it is defined in the following way:

\[
\delta_f = \frac{1}{m} \sum_{i} \left( \frac{Q^T_i - Q_i}{\delta Q_i} \right)^2
\]

where \( Q^T_i, Q_i \) are the desired target and current values of the \( i \)-th quantity of interest and \( \delta Q_i \) is the acceptable tolerance for that particular quantity. \( Q_i \) can represent any one of a multitude of properties of a multilayer and the merit function can be defined in terms of a combination of different types of \( Q_i \) because the use of \( \delta Q_i \) in the denominator normalizes all the diverse quantities [14]. However, most frequently \( Q_i \) represents \( T \) or \( R \) defined at different wavelengths, angles of incidence and planes of polarization.

An investigation to find which optimization method is the best for refinement of optical thin films not surprisingly did not reveal any one preferred routine – the best method depends not only on the particular problem but also on the stage in the optimization process [15]. Most thin film design programs provide a choice of several optimization methods.

**Synthesis**

One problem with optimization methods is that it helps to have a starting design with a performance that is a good first estimate of the required performance. For very complex problems choosing a good starting design is not an obvious task. Some people found good solutions to problems by refining several systems with an appropriate number of layers but with randomly selected thicknesses. Baumeister, for example, suggested that a suitable starting point for refinement would be a multilayer system composed of the type \((0.1H \ 0.1L)^N\), where \( H \), \( L \) correspond to quarter wave optical thicknesses of the highest and lowest refractive indices the available coating materials, and where \( N \) is large enough so that the overall thickness is sufficiently large for the problem on hand [16].

In the fifties and early sixties work was started in different laboratories on more powerful thin film design methods that did not require starting designs. Such computer programs are commonly called synthesis methods. In Russia, two groups published papers on such design methods [17,18]. Although it was easy to think of problems for which these methods would fail, they nevertheless inspired researchers at the NRCC to produce methods that would be more successful.

The **Comprehensive Search method** requires no starting design. In theory it finds, from a large number of combinations of layers with different refractive indices and thicknesses, that system which has a performance that is close to the global optimum solution to the problem [19]. This assumes that there is a solution that consists of a number of layers small enough so that the computation speed of the computer used is sufficient to find it in a reasonable time. Figure 3 shows the performance of an antireflection coating for the He-Ne laser wavelength (\( \lambda = 0.6328 \mu \text{m} \)) that reduced the substrate reflectance for unpolarized light to less than 1% for all angles of incidence up to 60° [20].

For problems that required many more layers for a solution, the **Gradual Evolution method** was proposed [19]. It was an extension of the comprehensive search method and it permitted solutions to much more complicated problems to be found. As already mentioned above, to find a solution to a complicated problem, the designer must have not only enough layers \( L \) at his disposal but also a sufficiently large overall optical thickness of the layer system. Because the solution is gradually evolved by adding a number of layers to the design in several stages, the solution found will no longer be close to the global optimum solution, but it can still consist of a reasonable number of layers. The example in Figure 4 is a linear filter in which the transmittance changes approximately from 1.0 to 0.0 within the 0.4–0.7 \( \mu \text{m} \) spectral region.

<table>
<thead>
<tr>
<th>Number of layers</th>
<th>Refractive index</th>
<th>Optical thickness A. Start</th>
<th>B. Refined</th>
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<tr>
<td>substrate</td>
<td>1.52</td>
<td></td>
<td></td>
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<tr>
<td>1</td>
<td>1.38</td>
<td>0.1500</td>
<td>0.2792</td>
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<tr>
<td>2</td>
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<td>0.1500</td>
<td>0.1630</td>
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<tr>
<td>3</td>
<td>1.38</td>
<td>0.1500</td>
<td>0.1477</td>
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<tr>
<td>4</td>
<td>2.30</td>
<td>0.1500</td>
<td>0.1506</td>
</tr>
<tr>
<td>5</td>
<td>1.38</td>
<td>0.1500</td>
<td>0.1509</td>
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<tr>
<td>6</td>
<td>2.30</td>
<td>0.1500</td>
<td>0.1440</td>
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<tr>
<td>7</td>
<td>1.38</td>
<td>0.1500</td>
<td>0.1434</td>
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<td>0.1500</td>
<td>0.1495</td>
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The refractive indices of these individual layers are then, one at a time, replaced by the refractive index of that of two or more pre-selected coating materials which yields the lowest value of the merit function. This process is very fast because during these calculations the optical thicknesses of the individual layers are not varied. The above procedure is repeated for all the layers of the system until the performance stabilizes. At that point adjacent layers with the same refractive index are merged and the system is refined to further improve its performance. The method can be extended to include absorbing layers [24]. In Figure 6 is shown an infrared black absorber filter that was designed by the Flip-Flop synthesis method.

Several people have described inverse Fourier transform methods for the design of optical thin film coatings. In their original implementations these methods could be only applied to problems in which the desired spectral reflectance or transmittance curve could be described by an expression that could be integrated [25-28]. The solutions were inhomogeneous layers that could be readily approximated by homogeneous layer systems. However, the need for an expression that could be integrated was rather limiting. At the NRCC a Numerical Inverse Fourier Transform method was developed that accepted the desired spectral curve in tabular form [29]. This was the first time that it was demonstrated that, for normal incidence of light, filters could be designed with transmission or reflection curves having any desired spectral profile, provided that the spectral region over which they were defined was reasonable. In Figure 7 is shown a filter designed by this method with a spectral reflectance curve that corresponds to the silhouette of the Parliament Buildings in Ottawa.

When refining an optical multilayer system, the process can come to a stop before a satisfactory solution has been obtained. This is because either the number of layers is not enough, or the overall thickness of the layer system is not sufficient. Originally Alexander Tikhonravov’s Needle method was designed to overcome this impasse [30]. In this method a very thin layer is inserted in the most effective position within a multilayer, in order to increase the number of construction parameters, so that the performance of the system could continue to be optimized through refinement. However, the needle method is much more than that. It is a true synthesis method that does not need a starting design. It is probably currently the most commonly used synthesis method in commercial thin film design software. Figure 8 shows a universal AR coating designed by the needle method which is effective on any glass substrate with a refractive index $n_s$ satisfying $1.48 < n_s < 1.75$ [31,32].
It should be mentioned, that even though such powerful design methods are now available, new thin film synthesis methods continue to be proposed. For example, only at the last Annual Meeting of the Society of Vacuum Coaters, a team from the Ecole Polytechnique in Montreal proposed the Refractive Index Step method, which the authors say, was inspired by the needle method (Figure 9) [33]. In it, an algorithm repeatedly finds the optimum position in the system where the introduction of a step in the refractive index of a layer will result in the largest improvement in the performance.

In addition to the above thin film synthesis programs for the design of optical multilayer coatings it is also possible to use various global minimum seeking programs that have been developed for the solution of general engineering problems [15]. Routines of this type that have been used in the past for thin film design include the Generalized Simulated Annealing-, the Monte Carlo Simulated Annealing-, the Revised Nelder-Mead Simplex- and the Genetic Algorithm methods. However, experience has shown, that even simple refinement methods, when used with a number of appropriate starting designs with random layer thicknesses, will likely yield satisfactory results more quickly.

**PROGRESS IN DEPOSITION TECHNOLOGIES**

To take full advantage of the enhanced power of the new design tools, improvements in two areas were crucial. One was the ability to deposit layers of different materials in a reproducible manner with properties that do not change in time. The other was the precise control of the thicknesses of the layers during the deposition, a subject that will be discussed in the next section.

The first of these two topics is not the main subject of this paper, but it is so important for the further development of the subject, that it has to be mentioned. In 1957 optical thin films were most commonly deposited from resistance-heated sources or by e-gun evaporation. Resistance heated sources had rather steady angular evaporation characteristics, which means that it was relatively easy to obtain uniform films by this process. But they were mostly useful for the deposition of fluorides, sulphides and other semi-conducting materials that evaporated or sublimed at relatively low temperatures. If the substrates could be heated during the deposition to sufficiently high temperatures, dense films were obtained whose properties did not change on exposure to the
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atmosphere. However, the above materials were relatively soft and not suitable for front surface use. Oxide films were much harder, but they required e-gun evaporation. As a rule, when deposited by this process, they resulted in porous layers that adsorbed water and changed their properties in air. Also, it was a challenge with this process to maintain good thickness uniformity over large areas.

In the last 50 years huge strides were made in this area. First, ion assisted e-gun evaporation resulted in denser coatings. Various magnetron-sputtering processes were developed that in addition to producing even denser coatings, made achieving good uniformity across large areas quite simple. Further, these processes are so steady and reproducible that for many applications, after suitable calibration, simple timing could be used to control the thicknesses of the layers. The speed of magnetron deposition is now approaching that of e-gun evaporation. Finally, ion-beam sputtering shows much promise for the future – coatings produced by this process have excellent properties, but the reproducibility of the deposition rate, the layer uniformity and system maintenance still need to be improved. Without the developments in deposition technology it would have been impossible to achieve the present state of the art in the manufacture of optical coatings.

PROGRESS IN THICKNESS CONTROL DURING THE LAYER DEPOSITION

After the spectacular demonstrations that thin film design methods can be used to find good solutions to quite complex filtering problems, much research was done to improve methods for the control of non-quarter wave layer systems.

At the NRCC a quarter wave layer monitoring technique was developed to control non-quarter wave layer thicknesses (Figure 10) [34]. This required the substrates to be stationary and the sources to be rotating. A monitoring glass (a) was placed next to the real substrate (b) that was in front of an adjustable rotating shutter (c) that could intercept a different fraction of the incident vapour for each layer. The resistance-heated sources (d) were mounted on a rotating commutator ring. This method worked very well for soft coating materials. But it was too difficult to adapt this method to e-gun evaporation of hard oxide layers.

The “most sensitive wavelength” monitoring method was then developed in which the monitoring wavelength for each layer was chosen by computer analysis so that the derivative of the transmittance with respect to thickness of the partial system including this layer had the highest possible value (Figure 1b) [34]. At this wavelength a given error in the transmittance corresponded to the smallest thickness error.

Many other groups proposed increasingly more sophisticated monitoring methods. The most significant contributions to this field probably came from Emil Pelletier’s group in Marseille. They published a number of papers that described various developments in wide-band optical monitoring in which the measured spectra of the gradually evolving system were compared to the calculated spectra of the system at the end of each layer. The deposition of the layer was terminated when the best fit between the two curves occurred. Figure 11 shows the calculated spectral transmittances for wide-band monitoring purposes of a two transmission peak band pass filter at different stages of its manufacture [35,36].

At this point it was not difficult to predict the future trend. Figure 12 shows the process envisioned at the NRCC in 1976 [20]. The starting point was a filter design that was optimized to be relatively insensitive to random thickness errors. It was assumed that the all-dielectric systems would be produced by a stable deposition process, that the optical constants of the coating materials were repeatable from run to run and that they were reliably determined by spectrophotometry or ellipsometry, and that the main unknowns would be the thicknesses of the layers. After the completion of each layer, the spectral transmittances would be measured and from these measurements the thicknesses of the layers deposited thus far determined. This step would be followed by the refinement of the thicknesses of the remaining layers of the system to compensate for the errors in the thicknesses of the layers produced thus far. This proposed deposition with real-time re-optimization process was first implemented at the NRCC in the early 1990-ties, after well-behaved and reproducible RF and AC magnetron sputtering sources became available. The process was completely automatic and, once initiated, it could proceed without supervision until the layer system was complete [37-39]. Figure 13 shows the target, design and measured spectral reflectances of a two-material 60-layer coating that has a spectral reflectance that corresponds to the silhouette of the Taj Mahal and that was designed by the needle synthesis method by Pierre Verly [38].

Figure 9. Design of a cut-off filter by the refractive index step method using a quarter-wave stack as a starting design [33].

Figure 10. Apparatus built at the NRCC for the use of the quarter wave monitoring method for the control of the thicknesses of non-quarter wave layers [34].

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Of course, significant contributions to such processes have also been made in many other laboratories [see, for example, references 40-42]. In particular, a combined team from the University of Rochester and the Moscow State University, headed respectively by Doug Smith and Alexander Tikhonravov, achieved promising results when they attempted to do the same with the much more complicated e-gun evaporation process [43].

It is obvious that even better results would be obtained if more accurate measurements could be made during the deposition process. A number of laboratories have pioneered the use of *in-situ* ellipsometric measurements which, in addition to the estimation of the thicknesses of the layers, permit the real-time determination of their optical constants [see, for example, references 42,44-47]. This is particularly important when the multilayer systems contain thin metal films – it is often crucial to determine accurately the extinction coefficients of the metal layers.

Several thin film design methods exist today that will provide the layer systems required to obtain almost any normal incidence spectral transmittance or reflectance for normal incidence of light, providing that the curve is specified over a reasonable range of wavelengths. However, computer methods are presently used for much more than just finding numerical solutions to a given design problem. They are now routinely used to perform various calculations such as the determination of the optical constants of coating materials and any inhomogeneities in deposited layers, the simplification of solutions through the removal of insignificant layers, calculations for the de-sensitization of layer systems to thickness and optical constant errors, random thickness and optical constants error analysis, the statistical modelling of monitoring strategies, the control of the deposition process, as well as the post-deposition evaluation of the layer systems. When all these processes are encompassed, it is indeed proper to talk about the “computer manufacturing” of optical multilayer coatings.

It should be stated at this point that a number of very good commercial thin film design software packages exist that provide such computational power [48,49,50]. Today it does not make any sense for a casual designer to write his or her own software.

The design of thin film filters for non-normal angles of incidence, especially when the filter is specified for both planes of polarization, can still be a challenge. Nevertheless, a number of innovative solutions for reflectors, polarizers, beam splitters and antireflection coatings (Figure 14), operating over a wide range of wavelengths and angles of incidence have been found in the past ten years.

In the past, from time to time, competitions have been held to test the state-of-the-art of thin film design [51-56]. More recently, Manufacturing Problems are being held at the OSA Topical Meetings on Optical Interference Coatings (OIC) that test the state-of-the-art of manufacturing. At the 2001 OIC meeting in Banff, participants were asked to design and produce a filter with spectral transmittance and reflectance curves that corresponded to the silhouette and its reflection in a lake of a mountain range in the Canadian Rockies [57]. This problem required non-absorbing coating materials and tested the ability of a laboratory to produce a coating with very complex T and R curves. In the best solution the experimentally produced curves on the average differed from the target values by only 0.98% (Figure 15).

At the 2004 OIC meeting in Tucson the manufacturing problem tested the ability of the participants to produce a 70:30 beam splitter for unpolarized light effective over a wavelength range of 0.45<λ<0.65 μm and operating at an angle of incidence of 60° (Figure 16) [58]. A number of very different solutions were submitted. In the best of these the average measured deviation from the target values in the wavelength range specified was only 0.79%. Another lesson learned during that exercise was that to make further progress in the manufacture of oblique incidence coatings, the accuracy of measurements at such angles has to be improved. It is very appropriate that measurement problems are being again held in order to focus on this topic [59-62].
At the 2007 OIC meeting, also to be held in Tucson, the manufacturing problem will test the ability to deposit accurate thin metal layers. The problem is to produce a coating that looks orange and blue in light reflected from the two surfaces, but appears grey in transmitted light.

Lastly, a forecast for the future. Let us suppose that a solution to a problem has been found that, for a certain set of materials and overall thickness, is a global minimum. Let us also assume that up to a certain point in the deposition process all the theoretical thicknesses of the layers have been deposited accurately, except that the thickness of the last layer has been exceeded. By re-optimization the thicknesses of the remaining layers of the system some of the damage caused by this error can be reduced. However, since the original solution was a global minimum, the performance of the original system cannot be completely recovered with the same number of layers. It follows that the only way to achieve the global minimum solution is not to make any errors.

The NRCC an ion beam sputtering system was used to make a filter with a spectral reflectance curve that followed the silhouette of the Palace of the Shoguns, in Kyoto (Figure 17) [63,64]. The departure of the theoretically calculated transmittance curve from the target value was 0.77%. To reduce this residual discrepancy still further, a more accurate measurement system would have to be installed. When the system was deposited without the real-time re-optimization of the remaining layers of the system, the average departure from the theoretical value was 3.27%. With re-optimization of the remaining layers of the system, the average departure was reduced to 1.28%. When the thicknesses that were overshot were etched down with the ion beam, the departure from the target value was only 0.77%. To reduce this residual discrepancy still further, a more accurate measurement system would have to be installed. This procedure will become practical once instrument manufacturers develop ion beam guns with more repeatable performance and less need for maintenance.

CONCLUSIONS

This article attempted to show that advances in computer technologies, when combined with the progress in deposition methods and thin film monitoring techniques, have resulted in the ability to manufacture complex optical multilayer systems with an accuracy that could hardly have been imagined 50 years ago. This, in turn, has led to a widespread use of optical coatings in a range of consumer-oriented products and sophisticated scientific and technological devices.

REFERENCES


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