Using fence post designs to speed the atomic layer deposition of optical thin films

Ronald R. Willey
Willey Optical, Consultants, 13039 Cedar Street, Charlevoix, Michigan 49720, USA
rwilley@freeway.net

Received 17 July 2007; accepted 12 September 2007;
posted 14 September 2007 (Doc. ID 85305); published 24 October 2007

Atomic layer deposition (ALD) at this time is much slower than conventional optical thin-film deposition techniques. A more rapid ALD process for SiO_2 has been developed than for other ALD materials. A fence post design for optical thin films has thin layers of high-index posts standing above a broad low-index ground. If a design for ALD can be predominantly composed of SiO_2 layers with thin high-index layers, the deposition times can be correspondingly shortened, and it is shown that the required performance can still be nearly that of more conventional designs with high- and low-index layers of equal thickness. This combination makes the ALD benefits of conformal coating and precise thickness control more practical for optical thin-film applications. © 2008 Optical Society of America

OCIS codes: 310.0310, 310.1860, 310.1620, 310.6805.

1. Introduction

Atomic layer deposition (ALD) or atomic layer epitaxy is generally the chemical vapor deposition of materials one monolayer at a time onto a surface. Such processes tend to be chemically self-limiting to one monolayer per cycle. The excess reactant is then purged from the chamber, and a second reactant is admitted to the chamber in which the surface is prepared for the next atomic thickness layer of the primary depositing species. The cycle times for a single monolayer are measured in several seconds, and the thicknesses per layer are measured in hundredths of a nanometer. Therefore the deposition rates are typically in hundredths of nanometers per second as compared to nanometers per second with physical vapor deposition (PVD). These ALD depositions are 1 or 2 orders of magnitude slower than the common PVD processes used in optical thin-film production. However, ALD does offer the great potential advantage of precise thickness control to within one atomic thickness and can produce uniform conformal coatings.

The semiconductor industry has expended extensive research and development effort over recent decades to take advantage of these characteristics, and most of the materials in the periodic table of the elements have been deposited by ALD, at least in the laboratory environment. One advantage for the semiconductor industry is the ability of ALD to uniformly deposit layers even in deep (i.e., 40:1) trenches. Examples can be seen when such trenches are completely filled with the ALD material [1]. The semiconductor industry has had a long experience with chemical vapor deposition (CVD), but those processes have tended to be at too high a temperature for most optical coating applications. More recent work using plasma-enhanced CVD at lower temperatures by researchers such as Martinu and Poitras [2] have made such techniques of greater possible application to optical thin-film production. Most of the current ALD processes seem to work well without plasma at temperatures less than the 300 °C processes that are common to optical coating. Most of the ALD processes are relatively easy to execute. Although the ALD process applications are relatively straightforward, the underlying chemistry can be complex. The field is greatly indebted to the chemical scientists who have studied these processes to understand the behavior of atoms and molecules on an individual and intimate basis, and they have found the precursors and process parameters that will produce the desired films of many materials.

0003-6935/08/13000C9-4$15.00/0

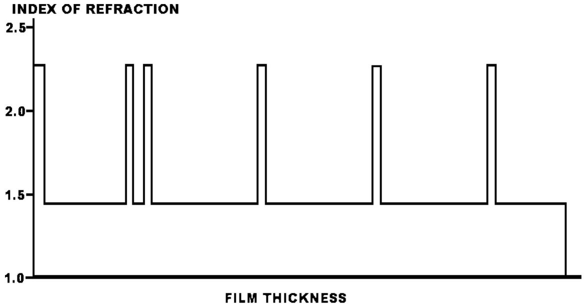

Fig. 1. This thin-film design appears to be fence posts of high-index layers above a low-index ground.

An unusual ALD process for the rapid deposition of SiO_2 has been reported by Hausmann *et al.* [3], where the process can be self-limiting at approximately 32 monolayers per cycle. This is a 1–2 orders faster deposition than for most other materials by ALD. This is more like the rates of optical thin-film PVD, and that forms the basis of this paper.

2. Fence Post Design Method

The concept of fence post (FP) designs was recently introduced [4]. The name fence post results from the appearance on an index versus thickness plot of narrow vertical posts of identical thicknesses of high-index layers above a ground of low-index layers as seen in Fig. 1.

The design of any narrow bandwidth minus or blocking filter by changing the ratio of the thickness between the high- and the low-index layers was recently reported [5]. The optical thickness of each pair of high- and low-index layers is the same as ordinary quarter-wave optical thickness (QWOT) designs at two QWOTs at the center of the blocking band. However, the width of the blocking band becomes less as the ratio of the overall thickness to the thinnest layer in the pair becomes greater than 2:1 (1 QWOT high index plus 1 QWOT low index). As the ratio is made higher, more layer pairs are needed to achieve a given optical density. A stack for a narrow blocker might be represented by (0.2H 1.8L)20 at the design blocking wavelength. The approach for more general spectral distributions would then be to hold the high-index layer thicknesses fixed and vary all the low-index thicknesses independently. This could be viewed as having all the same size fence posts and just varying the distance between them. This constrains the optimization to only half of the available variables, and that tends to make the designs somewhat more difficult for given requirements. The thin layers are not required to be all the same thickness, but that simplifying factor will be used here in most cases.

Index profiles similar to those in Fig. 1 have been employed by Schulz *et al.* [6,7] using PVD for hard, scratch-resistant antireflection (AR) coatings on plastic. In those cases, the thick low-index SiO_2 layers are separated by thin TiO_2 layers that are key to the hardness and scratch resistance. This same principle might be useful for other classes of coatings, and it has been shown [4] that essentially all classes of coatings can reasonably be approximated with the technique of identical thin layers spaced by thick layers of the opposite index.

If one were to design an optical coating for an ALD process using the rapid SiO_2 process for the low-index material and TiO_2 or HfO_2 for the high-index material, it would be advantageous to minimize the thickness of the high-index layers because they deposit at rates of 1 or 2 orders of magnitude slower than the low-index SiO_2 layers by the rapid process. Various designs using this FP concept are illustrated below.

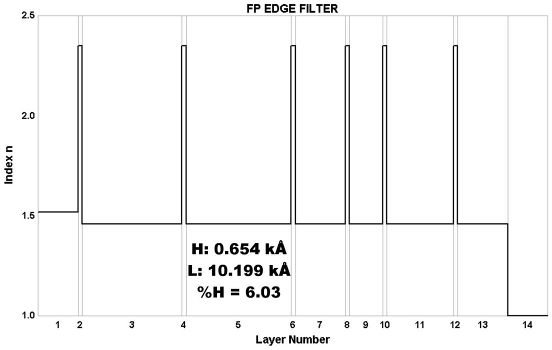

Fig. 3. Index profile of the 12 layer AR coating from substrate to air.

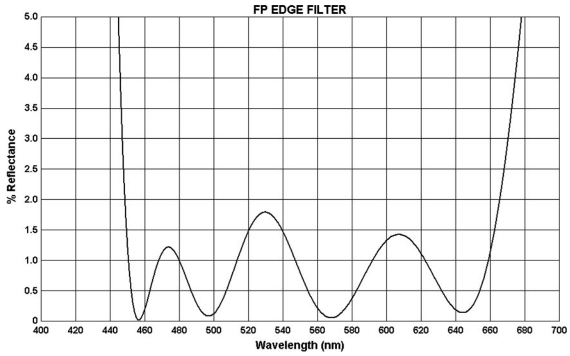

Fig. 2. Twelve (12) layer AR coating in which the high-index layers are all of the same thickness.

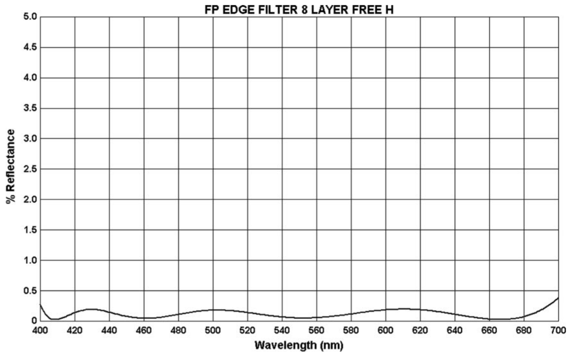

Fig. 4. Eight layer AR coating in which the high-index layers were allowed to vary for optimum results.

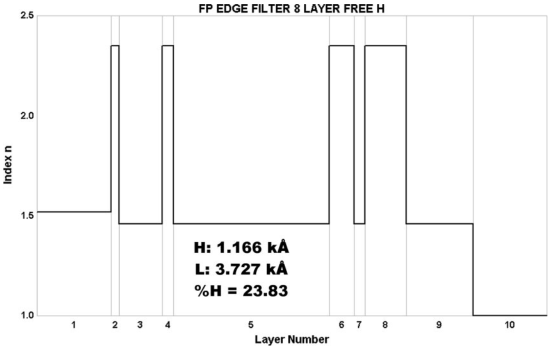

Fig. 5. Index profile of the eight layer AR coating from substrate to air.

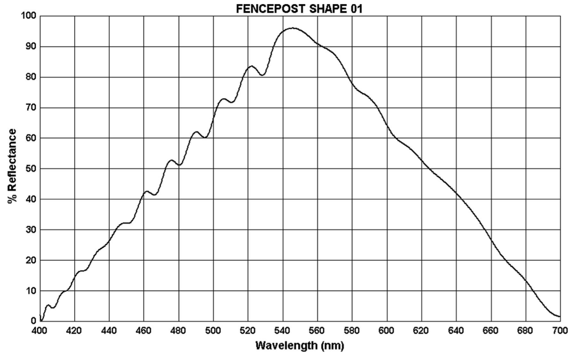

Fig. 8. General spectral shape of the FP design.

3. Possible Fence Post Designs

Reference [4] describes the use of FP design in more detail, but a few examples will be shown here to illustrate how much less high-index material thickness can be used in these designs. Figure 2 shows the spectral result of an AR coating design in which the thin high-index layers have been constrained to all have the same small thickness. This is seen to be too much of a constraint for any but the least demanding of AR requirements. However, when the high-index layers are allowed to vary in the design optimization, a much better design can be found as seen in Fig. 4. Figures 3 and 5 show the index profiles of these AR designs. Although the design of Fig. 4 has thicker high-index layers, the amount of high-index material is much less than the commonly used broadband AR designs. The index profile in Fig. 3 has 6.03% of its thickness in high-index material and that in Fig. 5 is 23.83%.

Edge filters and blockers can be designed with FPs as seen in Figs. 6 and 7. However, more layer pairs are needed with FPs to block or reflect a given bandwidth than would be required with conventional QWOT stacks. This could defeat the purpose of using FPs to speed up ALD processes. General spectral shapes such as those shown in Figs. 8 and 9 tend to be more amenable to FP designs. This particular one has about the same number of layers as its conventional counterpart.

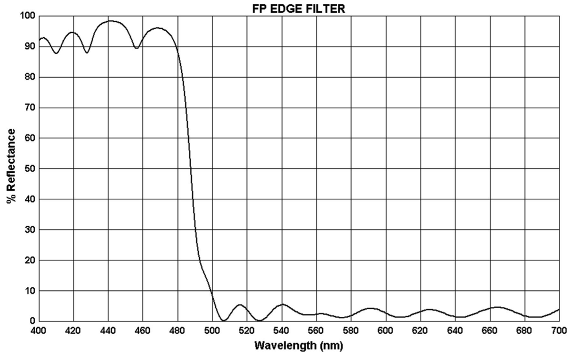

Fig. 6. Simple edge filter of the FP design.

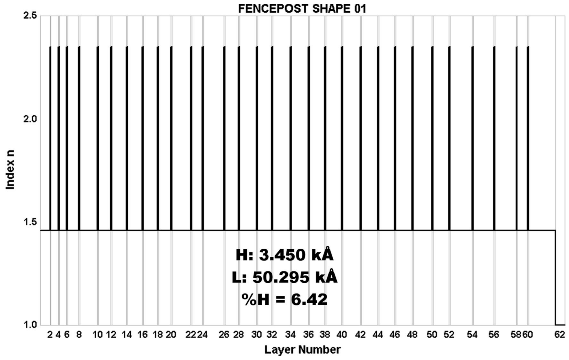

Fig. 9. Index profile of the general shape from substrate to air.

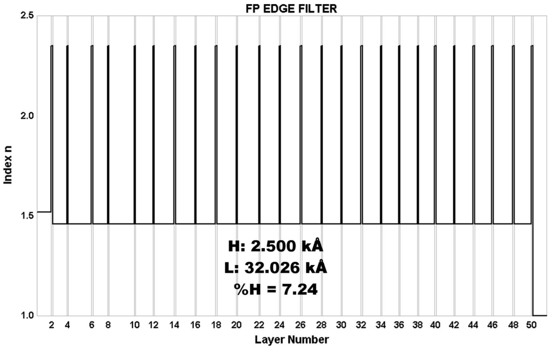

Fig. 7. Index profile of the edge filter from substrate to air.

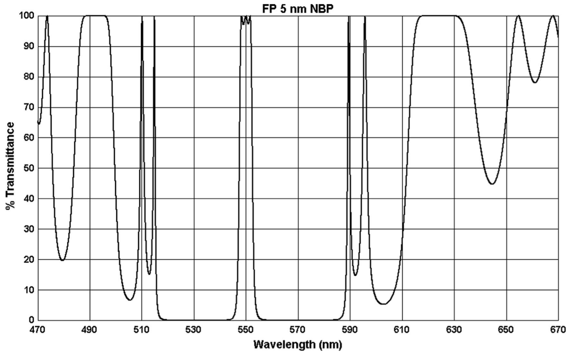

Fig. 10. NBP filter of the FP design.

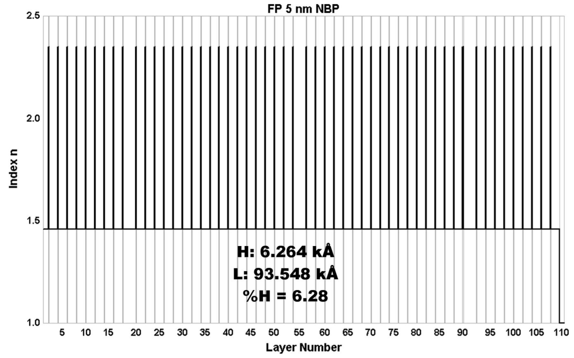

Fig. 11. Index profile of the NBP filter from substrate to air.

Narrow bandpass (NBP) filters tend to be among the more complex filters to produce. The FP design method can be used here also, as seen in Figs. 10 and 11. These do have some limitations that might be a hindrance in any given application. The blocking bands are not as wide or as deep as conventional QWOT designs. The narrower width could, however, be an asset if it were necessary to have passbands nearer the NBP region, such as seen in Fig. 10 at 495 and 625 nm. Other than the one AR design, all the above designs have only 6–7% of their overall thicknesses in high-index materials.

4. Conclusions

Designs have been illustrated for a variety of applications for which the thickness of high-index layers is 6–7% of the overall thickness and therefore almost an order of magnitude thinner high-index material than in conventional designs. This would then speed the ALD process by almost an order of magnitude for the high-index layers and more than an order of magnitude for the SiO_2 layers using the rapid process [3]. This should allow the practical entry of ALD into optical thin-film coating with ALD potential benefits of conformal coating and improved thickness control. Even if later process developments increase the deposition rates for high-index optical ALD materials, the fence post approach could still be beneficial for the various reasons described here and in the references.

References

1. J. J. Wang, X. Deng, X. Liu, A. Nikolov, P. Sciortino, F. Liu, and L. Chen, “Ultraviolet wave plates based on monolithic integration of two fully filled and planarized nanograting layers,” Opt. Lett. **31,** 1893–1895 (2006).
2. L. Martinu and D. Poitras, “Plasma deposition of optical films and coatings: a review,” J. Vac. Sci. Technol. A **18,** 2619–2645 (2000).
3. D. Hausmann, J. Becker, S. Wang, and R. G. Gordon, “Rapid vapor deposition of highly conformal silica nanolaminates,” Science **298,** 402–406 (2002).
4. R. R. Willey, “Design of optical thin films using the Fencepost Method,” Proc. Soc. Vac. Coaters **50,** 365–368 (2007).
5. R. R. Willey, “Design of blocking filters of any narrow bandwidth,” Appl. Opt. **46,** 1201–1204 (2007).
6. U. Schulz, U. B. Schallenberg, and N. Kaiser, “Antireflection coating design for plastic optics,” Appl. Opt. **41,** 3107–3110 (2002).
7. U. Schulz, U. B. Schallenberg, and N. Kaiser, “Symmetrical periods in antireflective coatings for plastic optics,” Appl. Opt. **42,** 1346–1351 (2003).