Optica Applicata, Vol. XVII, 4, 1987

Design and testing of AR coatings for NaCl optical elements used in CO_2 lasers

ANDRZEJ SZWEDOWSKI

Warsaw University of Technology, Institute of Design of Precise and Optical Instruments, ul. Karola Chodkiewicza 8, 02-525 Warszawa, Poland.

Three types of anti-reflection (AR) coatings for optical elements made of NaCl and used with high power CO_2 pulsed lasers were designed and tested. The coatings include one-layer AR film of NaF, two-layer ZnS/NaF coating being of AR type for $\lambda = 10.6$ μm and partly reflecting for $\lambda = 0.486$ μm, and two-layer AR coating of ZnSe/ZnS. The spectral characteristic and laser damage treshold were measured using 3 ns width pulse laser. Additionally, coatings durability was tested.

1. Introduction

Optical elements for high power CO_2 lasers used, for example, in plasma studies posses two basic characteristics: large diameter (sometimes more than 200 mm) and required resistance to laser radiation of high energy density in nanosecond pulses. These two factors and limited availability of other materials (for example, ZnSe) explain the choice of NaCl for windows, beam-splitting and Brewster plates. Low absorptance of order of 10^{-5} cm^{-1} and good damage resistance compensate such demerits as small hardness, brittleness and considerable solubility.

Optimization of performance parameters requires the application of surface coatings that improve the optical, mechanical and chemical characteristcs. In the Institute of Design of Precise and Optical Instruments one and two-layer coatings of NaF, and ZnS/NaF and ZnSe/ZnS, respectively, have been designed and tested. In the case of ZnS/NaF coating, besides requirements concerning the reduction of a reflection coefficient and increase of a resistance to humidity of the atmosphere, additional requirement of high reflection at $\lambda = 0.486$ μm should be fulfilled. It is dictated by the alignment process performed using argon laser.

The choice of materials for one layer AR coatings on NaCl for $\lambda = 10.6$ μm follows from low refraction index of the substrate, i.e., $n = 1.49$. In the case of $\lambda/4$ layers under the condition of permissible absorption not higher than 0.5 cm^{-1}, we can use [1]: NaF ($n = 1.23$), BaF_2 (1.40), SrF_2 (1.34) and ThF_4 (1.35). Sodium fluoride is the closest case to the optimal one. It is already used in high power laser systems [2]. The design of two and three-layer coatings provides greater flexibility in choosing refraction index especially when the condition of quantitized layer thickness for zero reflection can be relaxed. From Schuster's diagram we have the possibility of using several combinations such as ZnSe/ThF_4, ZnS/ThF_4, ZnS/BaF_2, ZnSe/BaF_2 or with the materials As_2S_3, As_2Se_3, SrF_2, TlJ, GATS.

Besides several demands typical to the materials used in thin film coating technology, additional ones are present. They follow from such requirements as low absorption, nonhigroscopicity of the last layer, good thermal conductivity and stability of characteristics of the film material (stechiometry). Materials satisfying these requirements are ThF_4, NaF, KCl, BaF_2, SrF_2, PbF_2, $KGeF_4$ and some of cryolites (excluding absorbing $NaAlF_4$), As_2S_3, As_2Se_3, ZnS, ZnSe, TlJ, GATS (Ge-As-Te-Se), Te-As-Se, and fluorides of rare earths. Materials, that cannot be used, contain H, He, Li, Be, C, N and O. The future material with high resistance to abrasion is diamondlike carbon (still difficulties concerning good adhesion to a subtrate [3] should be overcome), as well as polimer materials. Low resistance of NaCl to a humid atmosphere requires additional requirement of porosity – free layer, which should have glass or monocrystalic structure (as, for example, As_2S_3 [4]).

2. Testing of $\lambda/4$ NaF films for NaCl at 10.6 μm

Specific technology of AR coatings for NaCl at 10.6 μm is characterized by:
- higroscopicity of the substrate,
- nontypical preparation of a substrate surface before coating process,
- evaporation of a large volume of a material (about 20 times larger than for the visible region),
- long deposition time (50 min),
- damage threshold.

Preparation of a substrate for deposition differs from the typical one. For example, an elastic polymer layer is used for cleaning optical surfaces and protection of side surfaces of the element against the polishing powder. NaF, chosen because of the value of refraction index, exhibits good chemical affinity with substrate, 8 times lower solubility in water than NaCl, higher hardness (60 against 18.2, according to Knoop) and satisfactory laser radiation damage threshold. The latter one is almost equal to the one of uncoated NaCl. Because of a tendency of fluorides to create films of lowered density when deposited on cool substrate, it is necessary to warm up the substrate. On the other hand this material can tolerate rather low temperature gradients. This is why the temperature of 300°C was reached in 6 min. For the evaporation rate of 10 Å s^{-1} the deposition time is 40 min, and the whole process takes 3 hrs.

An influence of the substrate temperature on optical and mechanical properties of the coating was studied. The spectral transmittance of a plate with layers coated with a substrate heating to 300°C and without it was measured. Measurements conducted directly after finishing the process and after 8 months show a small decrease in transmittance. It is even smaller at $\lambda = 10.6$ μm for the plate with layers deposited on a heated substrate. For an unheated plate the decrease in transmittance was a considerable one (see Figs. 1 and 2). During 8 months the plates were stored in environmental conditions. Structured consistency of a layer (accompanied by higher density and, consequently, higher refraction index), being achieved for slower condensation of vapors of NaF, results in a smaller absorptance surface of molecules of H_2O. It causes a decrease in radiation absorptance to be observed near a characteristic band close to $\lambda = 6$ μm. The presence of water in upper layer of an optical element causes high absorptance of $10^3 cm^{-1}$, and lowers the damage

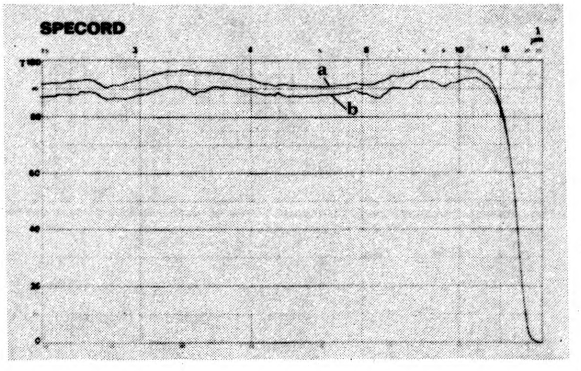

Fig. 1. Transmission of NaCl plate with anti-reflection NaF coating for $\lambda = 10.6$ μm: (a) directly after deposition; (b) 8 months after deposition. The layer was deposited on an unheated substrate

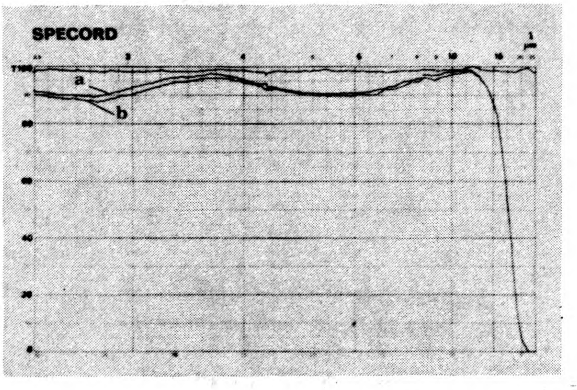

Fig. 2. As in Fig. 1, but for deposition on a substrate heated to 300°C

threshold value being, in general, inversely proportional to a square of a model layer thickness of H_2O.

Another factor that influences the layer structure, i.e., the deposition rate was studied. A spectral transmittance of samples with coatings obtained at 10 Å s^{-1} and 25 Å s^{-1} of a layer thickness growth, before and after a climatization trial (relative humidity of 90% and temperature 24.5°C during 18 hrs), was measured. A general decrease in transmittance for $\lambda = 2.5$ μm and $\lambda = 4$–6 μm was noted. It was not observed at 10.6 μm. An adhesion of layers to cold and warmed substrates to 100°, 200° and 300° was also investigated. "Scotch tape" was used for this purpose. It was found that about 20% und 10% of the surface was damaged in the case of unheated substrate to 100°, respectively. For the substrate heated to 200° and 300° no damage was observed. Some trials were also done to measure the force required to remove a special metal plate that was glued to the layer ("topple test"). However, no quantitative information was obtained because the glue layer (4 kinds) separated earlier from the film under study, or the layer was detached together with some parts of a substrate.

Laser radiation damage threshold was studied in an optical system shown in Fig. 4. A laser beam with half duration time of a pulse equal to 3 ns was focused on a

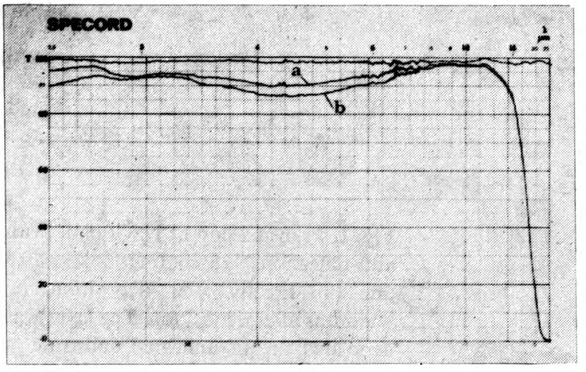

Fig. 3. Transmission of a plate with AR NaF coating (a) before, (b) after the humidity test

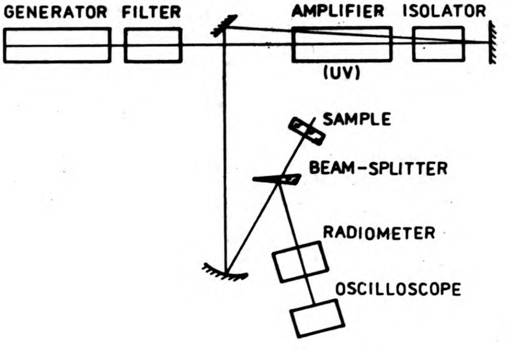

Fig. 4. Schematic representation of a setup for testing CO_2 laser damage threshold

back surface of a sample. For this purpose a spherical mirror of radius equal to 2000 mm was used. Statistical measurements showed that the damage threshold was greater than 3.4 ± 0.6 Jcm^{-2} under optimal technological conditions. For the layers obtained in a temperature 150°C it was equal to 1.9–2.3 Jcm^{-2}.

3. Studies of two-layer coatings

3.1. Zns/NaF coatings

In order to provide a possibility of laser system alignment, two-layer coatings were designed and tested. They are AR films for $\lambda = 10.6$ μm and partially reflective in visible range. The coatings are one quarter-wavelength layers of ZnS for $\lambda = 0.486$ μm and NaF for $\lambda = 10.6$ μm. The plane of polarization is perpendicular to the incidence plane. A calculated reflection coefficient for visible light is 0.11. The measured transmittance of whole plate with the second surface AR coated for $\lambda = 10.6$ μm was equal to 0.985 (a calculated one was 0.992). One of the problems to be solved during the deposition process was the selection of a substrate temperature to obtain good adhesion of NaF to NaCl. At the same time the stechiometry of ZnS should be avoided; it arises because of emission of Zn which increases absorptance. The coatings obtained showed to be resistant to abrasion and atmosphere humidity. The adhesion was slightly lower with comparison to NaF films. The same conclusion concerns a damage threshold (2.2 $\mathrm{Jcm^{-2}}$).

3.2. ZnSe/ZnS coatings

Antireflection two-layer coating of one quarter-wavelength thickness for $\lambda = 10.6$ μm does not give zero reflectance, but the transmittance of a plate with two-side deposited coatings should be equal, theoretically, to 0.985. Application of these materials was caused by a similarity of their properties, in particular, of the coefficient of linear expansion. The process was conducted with small deposition rate of approximately 5 $\mathrm{Å\ s^{-1}}$ taking into account an expected increase in absorptance proportional to the deposition rate of ZnSe. Because of that the deposition time was 45 min. Measurements of transmittance were performed directly after the process and after the "humidity trial" in a climatization chamber. Under the conditions stated above the spectral characteristic showed no change. The transmittance for $\lambda = 10.6$ μm was equal to 0.95. The layer under discussion is optimal from the point of view of protection against moisture of a fragile NaCl. An adhesion studied as a function of temperature showed the properties expected, i.e., for an unheated substrate the damaged area was about 50%, for the temperature of 150° it was about 2%. A damage is most frequently caused by the separation of ZnS layer from ZnSe film.

An influence of the substrate temperature was also noted when testing the laser radiation damage threshold. It is substantially lower for the coatings deposited on unheated substrate. For optimal temperature of 150°C the damage threshold was $1.4 \pm 0.1\ \mathrm{Jcm^{-2}}$. The coatings studied are designed for the use with medium energy density of a laser radiation as anti-reflection coatings of very good hydrophobic properties.

4. Final remarks

The work presented represents a part of activity in the field of technology of optical elements used with high energy infrared radiation. The laser damage threshold can be increased by further optimalization of the process of preparation of the substrate with minimum surface absorption and the deposition process itself. Further optimalization of the performance properties of multi-layer coatings is the more possible because of the possibility of application of other film materials.

Acknowledgements — The results presented in this work were obtained with cooperation of Eng. S. Witek of the Institute of Design of Precise and Optical Instruments of the Warsaw University of Technology and Z. Sikorski, M. Sc., of the Institute of Plasma Physics and Laser Microsynthesis.

References

[1] SPARKS M., NBS Special Publication No. 462 (1976), 203.
[2] NEWMAN B. E., Opt. Eng. **18** (1979), 579.
[3] BENNETT H., Appl. Opt. **22** (1983), 3276.
[4] YOUNG P. A., Thin Solid Films (1970), 423.

Received June 17, 1987

Разработка и исследование просветляющих покрытий на оптических деталях, предназначенных для CO_2 лазеров

Разработаны и исследованы три оптические тонкоплёночные покрытия на поверхностях оптических деталей из NaCl, предназначенных для CO_2 лазеров большой мощности. Эти просветляющие плёнки из NaF, ZnSe/ZnS и плёнка ZnS/NaF просветляющая при $\lambda = 10.6$ μm и частично рефлексная при $\lambda = 0.486$ μm. Исследовано спектральное пропускание, порог лазерного разрушения для длительности импульса 3 ns и прочность покрытий.