Optica Applicata, Vol. IX, No. 4, 1979

Ellipsometric examination ef anodized aluminium layers within the visible spectral range*

Elżbieta Idczak, Ewa Oleszkiewicz

Institute of Physics, Technical University of Wrocław, Wrocław, Poland

The system of Al_2O_3 on Al layers obtained by the aluminium anodization method in the visible spectral range 450–650 nm have been subjected to ellipsometric examinations. The ellipsometric angles Δ and Ψ have been measured both for the aluminium layers and the Al_2O_3 on the Al substrate. On the base of the ellipsometric measurements the optical constants for aluminium layers have been calculated by a numerical method, taking into account the existence of a natural oxide ** layers as well as the refractive index and different thicknesses of the aluminium oxide layers deposited on aluminium. It has been stated that the refractive index of the aluminium oxide on aluminium layers does not depend practically upon the oxide thickness and that the aluminium layers exhibit the normal dispersion within the visible spectral range.

Introduction

The amorphous Al_2O_3 films on Al layers have been frequently exploited in microelectronics and other branches of technology. The optical properties of such layer system may be examined by ellipsometric method, which has been more and more frequently used in investigations of thin films deposited on all kinds of substrates [1–11]. The Al_2O_3 layers on Al were examined carefully [1–5, 10–16] but all these investigations concerned only one wavelength 546.1 nm or 632.8 nm. Hence, the examination of these layers within the whole visible spectral range 450–650 nm seemed to be of some interest.

Thus, the purpose of this paper was to examine Al_2O_3 films on Al layers for different Al_2O_3 layer thicknesses with the help of the ellipsometric method within the 450–650 nm spectral range.

Experimental results of investigations

The examined Al_2O_3 films on Al layers were prepared in the following way: the Al layers with thicknesses of the order of 1200 nm were evaporated in 1.33 mPa vacuum on polished and carefully cleaned BK-7 glass plates from a tungsten heater with the 40 nm/s rate.

---

* This work was carried on under the Research Project M.R. I. 5.

** The oxide layer produced under the influence of the atmosphere.

Al layers have been then anodized in order to obtain the aluminium oxide; anodization being carried out at constant current density in the non-aqueous electrolyte (in the form of ammonium pentaborate dissolved in ethylene glycol). The electrolyte temperature was kept at 358 K. The cathode was a platinum electrode with the surface much greater than the anode surface. To obtain Al_2O_3 layers of different thicknesses different values of threshold voltage (2.5–100 V) have been applied.

All the ellipsometric measurements were performed with the ellipsometer described in [17]. For an accurate determination of the ellipsometric angles Δ and Ψ the azimuths of the polarizer and the analyser at crossed position were measured in two zones; the azimuth of the compensator (a quarter-wave plate) was constant and amounted to $3/4\pi$.

From the indications of the analyser and the polarizer settings the ellipsometric angles Δ and Ψ were determined on the basis of the accurate ellipsometric formulae [18, 19]. The measurements of and Ψ have been performed for two angles of incidence 70° and 75° in the visible spectral range.

Discussion of the results

In order to determine the refractive index and thickness of a dielectric layer on an absorbing substrate from a single ellipsometric measurement, i.e. from the measurement of the angles Δ and Ψ it is necessary to know the optical constants of the substrate. If an opaque layer is used as the substrate of the layer system, then the optical constants of such a substrate depend essentially upon the technological conditions and the measurement method applied [12, 13].

In order to determine the optical constants of Al layers serving as substrates for Al_2O_3 layers the aluminium layers were measured ellipsometrically after evaporation and before anodization within the whole 450–650 nm spectral range. When calculating the optical constants of aluminium the existence of the natural oxide layer of 4–5 nm thicknesses [8], which appears due to the atmosphere action on freshly evaporated Al layer, has been taken into account.

In the calculations the assumed value 1.62 of the index of refraction of the natural oxide layer and its thickness (5 nm) were taken from the literature [8]. Since the value of refractive index of Al_2O_3 obtained from the experiment appeared to be greater than 1.62 we have tried to calculate the optical constants of the substrate by assuming the value of the refractive index of aluminium oxide layer to be 1.74 and its thickness equal to 5 nm. The results obtained for optical constants of aluminium layers – when taking into account the oxide layer produced under the influence of the atmosphere – have been shown in fig. 1, from which it follows that the ignoring of oxide layer results in too low estimation of

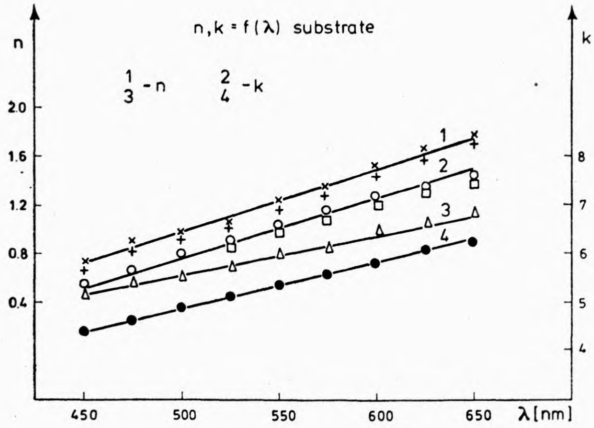

Fig. 1. The spectral dependence of the optical constants (n, k) for opaque aluminium layers:

● △ — optical constants of the aluminium layers, the natural oxide layer being not taken into account; + □ — optical constants of aluminium layers considering the presence of intrinsic oxide layer with $n_1 = 1.62$, $d = 5$ nm; × ○ — optical constants of the aluminium layers considering the presence of intrinsic oxide layer with $n_1 = 1.74$, $d = 5$ nm

both the refractive index (n) and the absorption coefficient (k) calculated from the ellipsometric measurements. Besides, the optical constants (n and k) of the aluminium layers proved to be insensitive to the accepted value of the refractive index of the natural oxide. The results obtained for the optical constants of aluminium layers (fig. 1) within the visible spectral region are practically the same, regardless whether the accepted value of the refractive index of the aluminium oxide is 1.62 or 1.74.

Next the aluminium layers have been subjected to anodization (during some time) until the definite voltage is achieved, the latter being dependent upon the thickness of the obtained aluminium oxide layer. For the obtained Al_2O_3 on Al layers system the ellipsometric angles Δ and Ψ were measured within the visible spectrum region. The results of ellipsometric measurements vs. the aluminium oxide layer thickness are shown in figs. 2–7 for several wavelengths of light. The measurements start at the point which represents the non-anodized aluminium layers and as the oxide layer thickness grows the measurement points are shifted along the ellipsometric curve in the counter-clock-wise direction. We tried to match the theoretical ellipsometric curve to the experimental measurements $\Delta = f(\Psi)$ by assuming different values of the refractive index of the aluminium oxide layer. The best consistency of the experimental results with the theoretically calculated ellipsometric curve was obtained for the refractive index of the aluminium oxide equal to 1.8. We did not manage to match the theoretical curve to the experimental results for the thickest layers of the order of 100 nm which might indicate some heterogeneity of the aluminium oxide layers obtained by anodization.

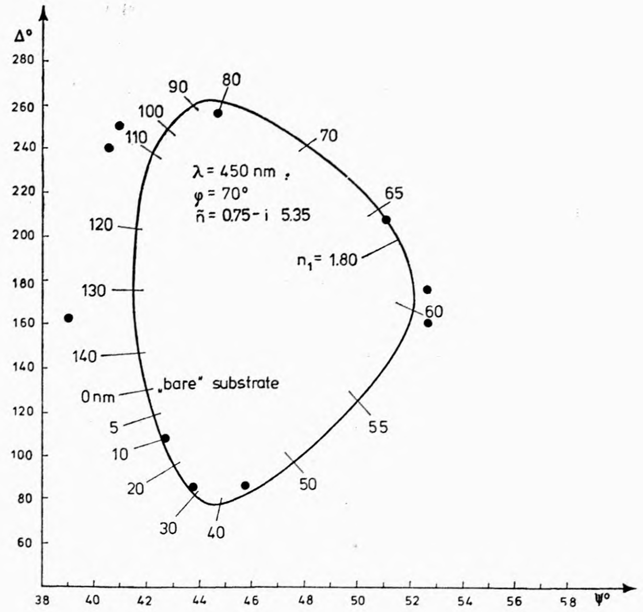

Fig. 2. Experimental ellipsometric measurements $\Delta = f(\Psi)$ for the anodized aluminium layers ($\varphi = 70°$, $\lambda = 450$ nm, $n = 0.75$, $k = 5.35$).

Full curve is calculated theoretically ($n_1 = 1.80$, the numbers denote the thicknesses of the aluminium layers, ○ – the experimental results)

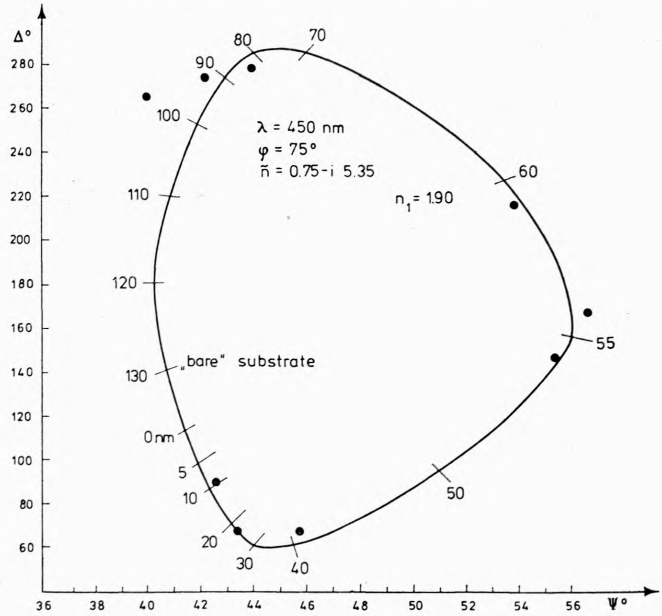

Fig. 3. Experimental ellipsometric measurements $\Delta = f(\Psi)$ for the anodized aluminium layers ($n = 0.75$, $k = 5.35$) for $\varphi = 75°$ and $\lambda = 450$ nm.

Full curve is calculated theoretically ($n_1 = 1.90$, the numbers denote the thicknesses of the aluminium oxide layers, ○ – the experimental results)

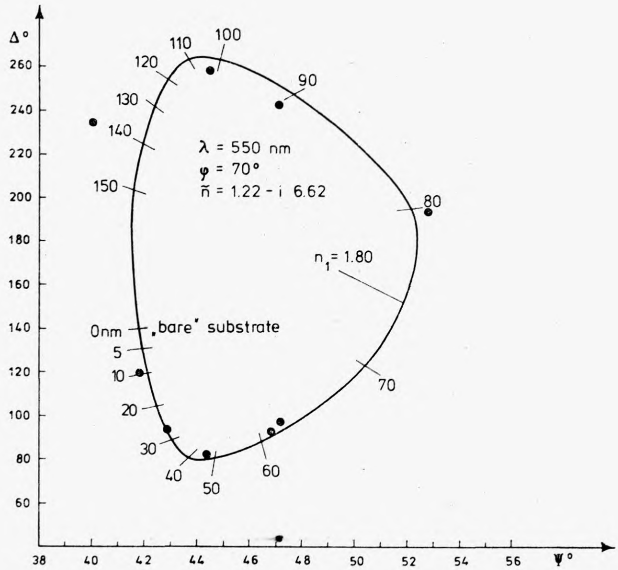

Fig. 4. Experimental ellipsometric measurements $\Delta = f(\Psi)$ for the anodized aluminium layers ($\varphi = 70°$, $\lambda = 550$ nm, $n = 1.08$, $k = 6.33$)

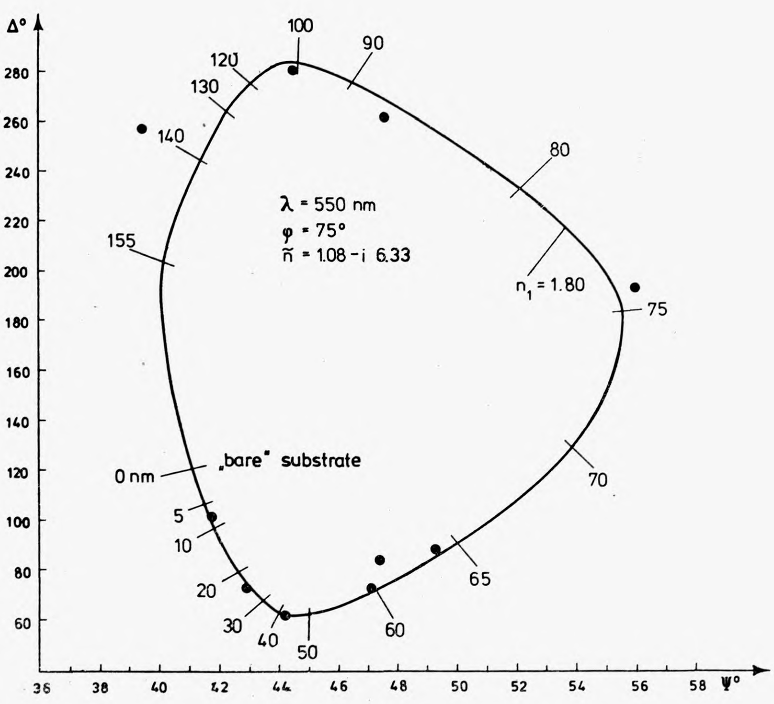

Fig. 5. Experimental ellipsometric measurements $\Delta = f(\Psi)$ for the anodized aluminium layers ($\varphi = 75°$, $\lambda = 550$ nm, $n = 1.08$, $k = 6.33$)

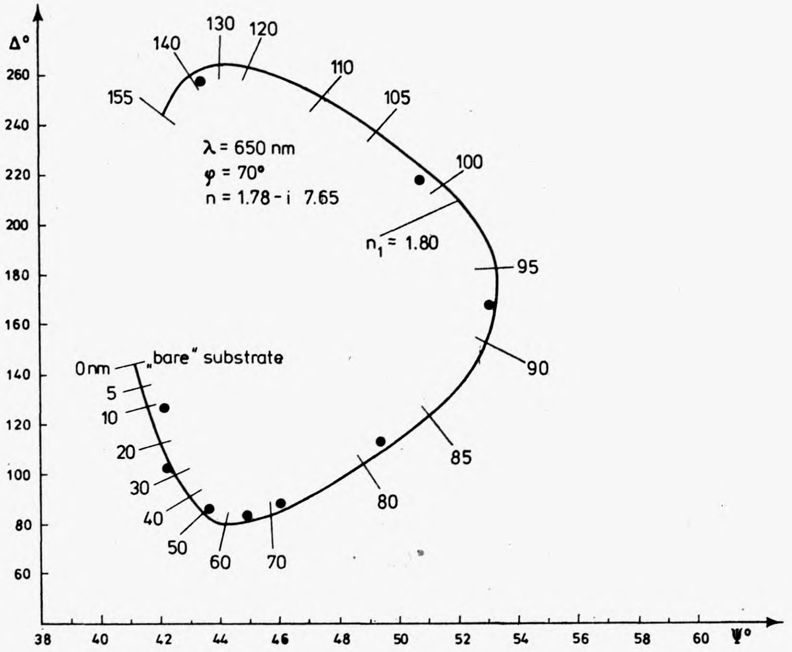

Fig. 6. Experimental ellipsometric measurements $\Delta = f(\Psi)$ for the anodized aluminium layers ($\varphi = 70°$, $\lambda = 650$ nm, $n = 1.78$, $k = 7.65$)

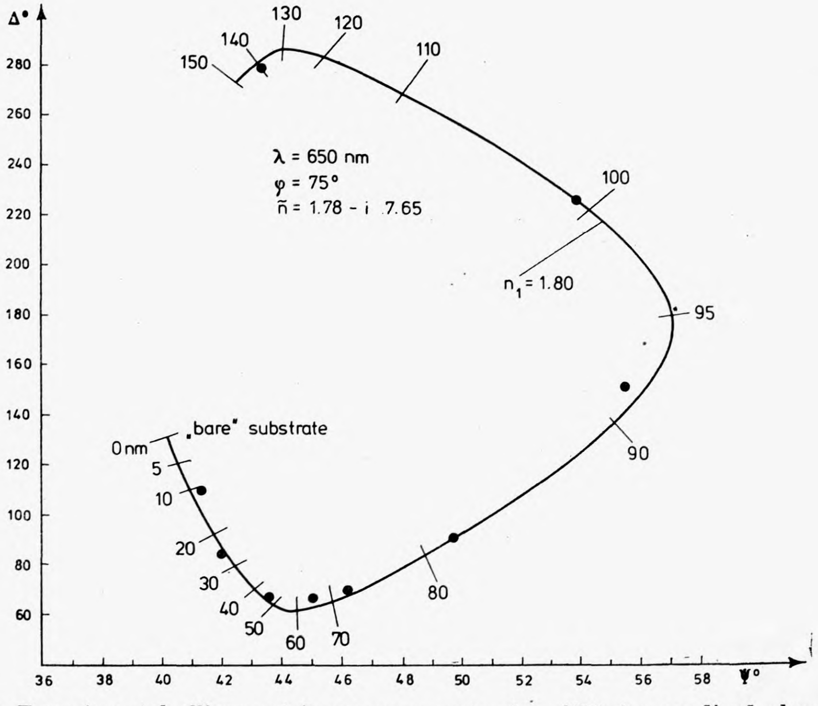

Fig. 7. Experimental ellipsometric measurements $\Delta = f(\Psi)$ for anodized aluminium layers ($\varphi = 75°$, $\lambda = 650$ nm, $n = 1.78$, $k = 7.65$)

The fact that the refractive index of Al_2O_3 changes with the increase of anodized layer thickness was reported in papers [15, 16]. Assuming, however, the aluminium oxide-on-aluminium layers to be uniform we have calculated the effective refractive index and thickness of the oxide layer for nine wavelengths from the 450–650 nm region and for two of angles incidence, 70° and 75°, of the light beams. The calculations were carried out with the help of a specially developed programme for the computer [20]. The results obtained for the refractive index of the aluminium oxide-on-aluminium layers are shown in figs. 8–10 as functions of

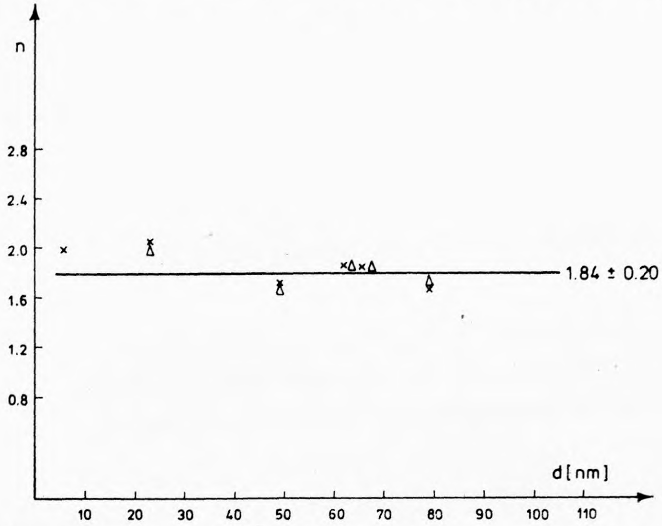

Fig. 8. Dependence of the refractive index on the thickness of the aluminium oxide layers deposited on aluminium, $\lambda = 450$ nm,
× – parameters of the natural oxide layer $n_1 = 1.62$, $d = 2$ nm; △ – $n_1 = 1.62$, $d = 5$ nm

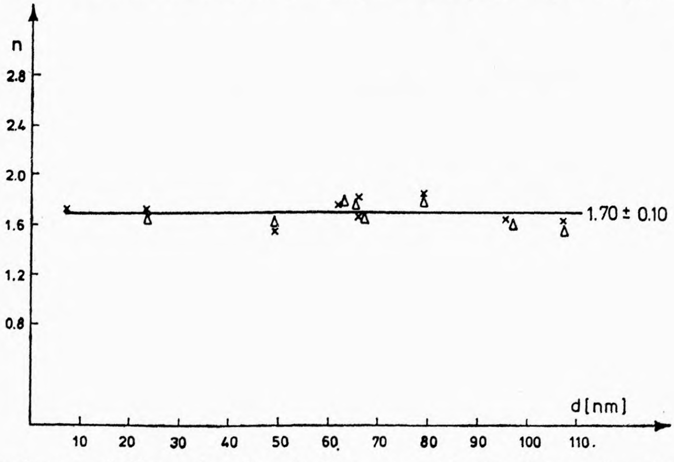

Fig. 9. Dependence of the refractive index upon the thickenss for the aluminium oxide on aluminium layers $\lambda = 550$ nm:
× – natural oxide layer with $d = 2$ nm and $n_1 = 1.62$; △ – natural oxide layer with $d = 5$ nm and $n_1 = 1.62$

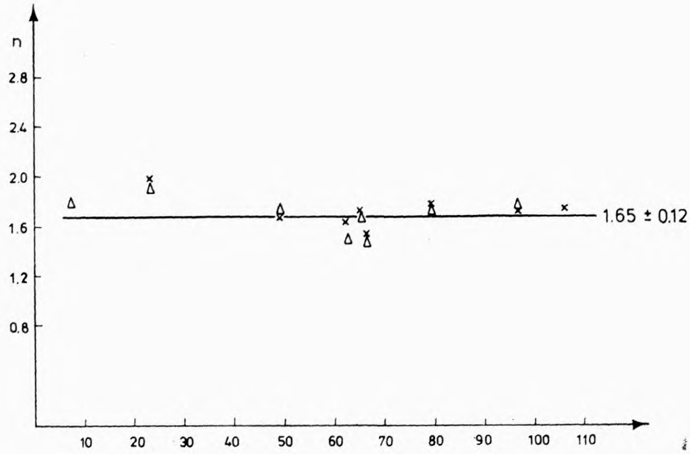

Fig. 10. Dependence of the refractive index upon the thickness for the aluminium oxide on aluminium layers $\lambda = 650$ nm,

× – natural oxide layer with $d = 2$ nm and $n_1 = 1.62$; △ – natural oxide layer with $d = 5$ nm and $n_1 = 1.62$

the oxide layer thickness for three wavelengths. As it follows from these figures the refractive index of the aluminium oxide layers obtained by anodization method does not depend, practically, upon the layer thickness and for $\lambda = 55$ nm it amounts to 1.70 ± 0.10.

The spectral dependence of the refractive index of the aluminium oxide layers of thickness $d = 62$ nm has been shown in fig. 11 from which it may be seen that the aluminium-oxide-on-aluminium layers exhibit the normal dispersion within the spectral range.

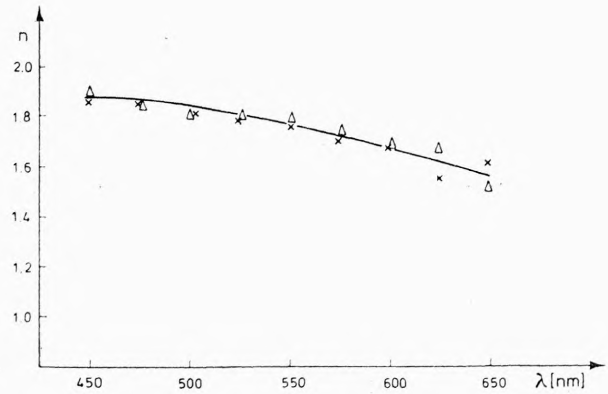

Fig. 11. Dispersion $n(\lambda)$ for the aluminium oxide on aluminium layers with $d = 62$ nm

× – natural aluminium oxide layer with $d = 2$ nm and $n_1 = 1.62$; △ – natural aluminium oxide layer with $d = 5$ nm, and $n_1 = 1.62$

Conclusions

Some results obtained in this work may be compared with the data concerning the aluminium oxide layers obtained by other authors. For the wavelengths 546.1 and 632.8 nm the values of the refractive index of the aluminium oxide layers produced in our laboratory are consistent within the experimental error with the literature data. From the examinations carried out for the Al_2O_3 on Al layer substrate the following conclusions may be drawn:

1. To obtain the correct values of the refractive index and the thickness of the surface layers the values of the optical constants of the substrate must be known accurately. If – in case of aluminium – the existance of a natural oxide layer is neglected, the estimated values of the refractive index and the absorption coefficients of the aluminimu layer are too low.

2. If the existence of the natural oxide layer is taken into account it is sufficient to know only an approximate value of the refractive index of this layers e.g. 1.72 ± 0.10 for aluminium, since the optical constants are not much sensitive to the values of the refractive index of the surface layer.

3. The aluminium oxide layers exhibit the normal dispersion within the visible spectral range, while the refractive index of those layers does not depend practically upon their thicknesses within the interval 6–110 nm*

Acknowledgements – The authors express their best thanks to Doc. Dr. S. Osadnik and Mgr Ing. Z. Znamirowski for making anodization technology available and for the assistence during anodization the aluminium layers, and to Prof. C. Wesołowska for the interest shown to this work.

References

[1] Shklyarevskii J. N., El Shagly A. F. A., Idczak E., Sol. St. Commun. **9** (1971), 1737.

[2] Shklyarevskii J. N., El Shagly A. F. A., Idczak E., Opt. i Spektr. **33** (1972), 1157.

[3] Shklyarevskii J. N., El Shagly A. F. A., Jarovaya R. G., Kostyuk V. P., Sol. St. Commun. **10** (1972), 1045.

[4] Shklyarevskii J. N., El Shagly A. F. A., Jarovaya R. G., Kostyuk V. P., Opt. i Spectrosk. **36** (1974), 199.

[5] Idczak E., Żukowska K., Optica Applicata **VIII** (1978), 55.

[6] Barret M. A., [In:] *Ellipsometry in the Measurements of Surface and Thin Films*, Ed. E. Passaglia, R. R. Stromberg, J. Kruger, Washington 1964.

[7] Goldstein R. M., Lederich R. J., Leonhard F. W., J. Electrochem. Soc. **117** (1970), 503.

---

* This paper has been presented at the Fourth Polish-Czechoslovakian Optical Conference in Rynia (near Warsaw), September 19–22, 1978.

[8] Fane R. W., Neal W. E., J. Opt. Soc. Am. **60** (1970), 790.
[9] Badia M., Thin Sol. Films **13** (1972), 329.
[10] Sherichun F., Rowe E. C., J. Appl. Phys. **41** (1970), 4128.
[11] Jegorova G. A., Potrapov E. V., Rakov A. V., Opt. i Spektrosk. **41** (1976), 643.
[12] Hass G., Waylonis J. E., J. Opt. Soc. Am. **51** (1961), 719.
[13] Ehrenreich M., Philipp H. R., Sedall B., Phys. Rev. **132** (1963), 1920.
[14] Dell'Oca J., Thin Solid Films **26** (1975), 371.
[15] Dell'Oca J., Fleming P. J., J. Electrochem Soc. **123** (1976), 1487.
[16] Young L., *Anodic Oxide Films*, Academic Press, New York 1961.
[17] Idczak E., Oleszkiewicz E., Żukowska K., Report No. 249, Institute of Physics, Technical University of Wrocław, Wrocław 1976.
[18] Smith P. H., Surface Sc. **16** (1969), 34.
[19] Gorshkov M. M., *Elipsometriya*, Izd. Sovet. Radio, Moskva 1974.
[20] Idczak E., Oleszkiewicz E., Żukowska K., Optica Applicata **IX** (1979).

Received, September 30, 1978

Эллипсометрические исследования анодированных слоёв алюминия в видимой области спектра

Проведены эллипсометрические исследования расположения слоёв Al_2O_3 на Al, полученные методом анодирования в видимой области спектра 450–650 нм.

Измерены эллиптсометрические углы Δ и Ψ как для слоёв алюминя, так и для расположения слоёв Al_2O_3 на Al. На основе эллипсометрических измерений с помощью численного метода были рассчитаны оптические константы слоёв алюминия, учитывая в расчётах слой природного окисла, а также коэффициент преломления и толщину слоёв окиси алюминия различной толщины на алюминии. Выявлено, что коэффициент преломления слоёв окиси алюминия на алюминии практически не зависит от толщины слоя окиси, а также, что слои окиси алюминия в видимой области спектра имеют нормальную дисперсию.