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EFFECT OF NEUTRON IRRADIATION ON THE PROPERTIES OF FABRY-PEROT INTERFERENCE FILTERS

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The effect of reactor neutron irradiation on the optical properties of ZnS — cryolite Fabry-Perot interference filters was investigated. Optical transmittance spectra of the filters were taken before and after irradiation; neutron fluxes in the range of 10^{13}—10^{18} neutrons/cm^2 were used. The shift of the peak transmittance wavelength as well as the decrease in transmittance were observed for neutron fluxes higher than $\sim 10^{17}$ neutrons/cm^2. The change in the peak transmittance wavelength was related to the change in optical thickness of the layers, i.e. to the change in refractive index and geometrical thickness induced by neutron irradiation. The decrease of the density as well as of the index of refraction of ZnS layers was calculated using the experimental results and a simple displacement theory.

1. *Introduction*

The influence of energetic nuclear irradiation on the properties of dielectric narrow-band transmittance interference filters has been, scarcely, investigated. In the present work the changes in the optical transmittance spectra of ZnS — cryolite Fabry-Perot interference filters caused by reactor neutron bombardment were studied.

2. *Experimental*

Filters were formed by vacuum deposition of zinc sulphide and cryolite (Na_3AlF_6) onto unheated suprasil substrates. The filter design was:

group *A* filters — suprasil 4(HL) 8H 4(LH) air, and

group *B* filters — suprasil 4(HL) 64H 4(LH) air,

where H and L denote the quarter-wave thickness of ZnS and cryolite, respectively. Filters were covered with a plate of suprasil coverglass stuck at the edges with polystyrene in benzene in order to protect them from water vapour sorption. The samples were mounted in aluminium containers for irradiation in the central channel of 250 kW TRIGA Mark II reactor of the »Jožef Stefan« Institute, Ljubljana. Neutron fluxes were used in the range of 10^{13}—10^{18} neutrons/cm^2. Optical transmittance spectra were taken before and after irradiation by means of a Zeiss monocromator. Suprasil substrates alone, without covering, were subjected to irradiation and their transmittance spectra were measured before and after neutron bombardment.

3. *Results and discussion*

It was observed that neutron bombardment led to a shift of the peak transmitlance wavelength of filters towards shorter wavelengths when neutron fluxes are targer than $\sim 10^{17}$ neutrons/cm^2. Fig. 1 shows a shift, $\Delta\lambda$, of the maximum transmittance wavelength versus neutron flux for filters of group *A* (curve A), as well as of group *B* (curve B); in both cases, $\Delta\lambda$ is about zero below a flux of 10^{17} neutrons/

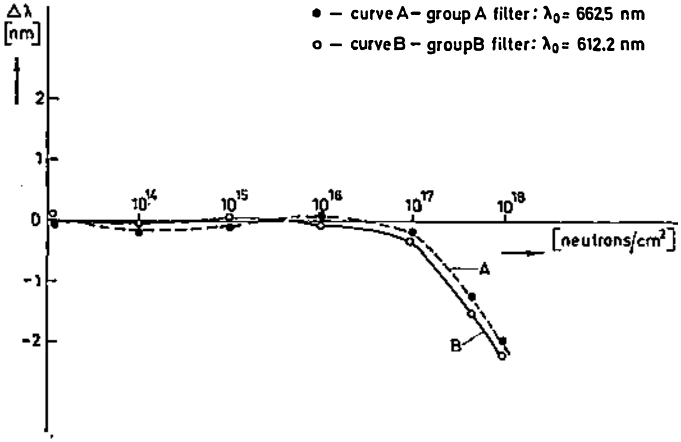

Fig. 1. Shift $\Delta\lambda$ of peak transmittance wavelength vs. neutron flux; curve *A*: filter belonging to group *A* filters, and curve *B*: filter belonging to group *B* filters.

cm^2 and then it increases towards negative values. As well as this shift, transmittance spectra also show a broadening and a splitting of the initial peak into two peaks, with the distance $\Delta\lambda'$ between them (Fig. 2).

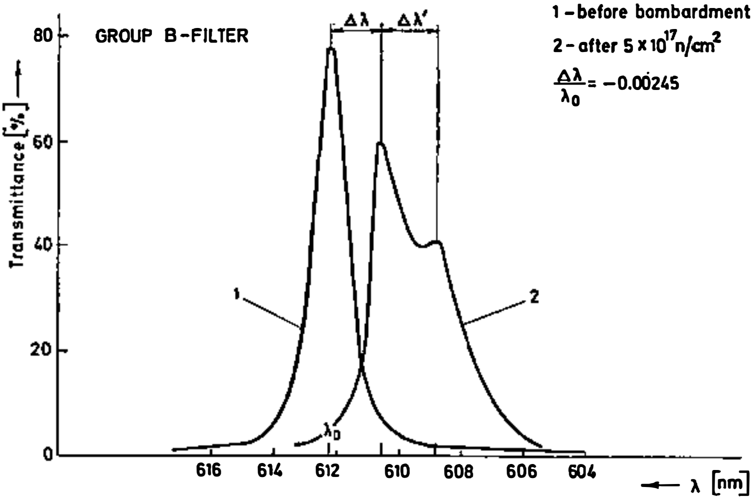

Fig. 2. Transmittance band of a filter belonging to group B filters: curve 1 — before irradiation; curve 2 — after $5 \cdot 10^{17}$ neutrons/cm^2. Initial peak transmittance wavelength $\lambda_0 = 612.2$ nm; relative change of the peak transmittance wavelength after $5 \cdot 10^{17}$ neutrons/cm^2 is, $\frac{\Delta\lambda}{\lambda_0} = -0.00245$.

Let us discuss the shift $\Delta\lambda$ of the maximum of the original transmittance band. It is necessary to point out that in these irradiation experiments suprasil substrates had no influence on the behaviour of the filters. Suprasil substrates alone were exposed to the corresponding neutron fluxes; no changes in the optical transmittance spectra before and after bombardment were observed. The decrease in peak transmittance wavelength of the filter due to neutron irradiation might presumably be connected with the decrease in optical thicknesses of the layers. Furman[1] showed that the relative change $\frac{\Delta\lambda}{\lambda_0}$ (λ_0 is the initial peak transmittance wavelength) of a dielectric narrow-band transmittance interference filter with symmetrical reflectors is related to the relative change of optical thicknesses of the layers with high and low index of refraction:

$$\frac{\Delta\lambda}{\lambda_0} = B_1 \frac{\Delta h_h}{h_h} + B_2 \frac{\Delta h_l}{h_l}, \tag{1}$$

where h denotes the optical thickness of the layer ($h = n \cdot d$, n is the index of refraction and d is the geometrical thickness), and the subscripts h and l denote the high and low index of refraction layers, respectively. In our case, with an even number of layers in the reflectors, the coefficients B_1 and B_2 are defined in the following way1):

$$B_1 = 1 - B_2,$$

$$B_2 = \frac{n_h n_l}{(p + B)(n_h^2 - n_l^2)} \tag{2}$$

p is the order of the filter and $B = \frac{n_l}{n_h - n_l}$. The relative changes of optical thicknesses can be written in the form:

$$\frac{\Delta h_h}{h_h} = \frac{\Delta n_h}{n_h} + \frac{\Delta d_h}{d_h},$$

$$\frac{\Delta h_l}{h_l} = \frac{\Delta n_l}{n_l} + \frac{\Delta d_l}{d_l}. \tag{3}$$

Taking into account relations (2) and (3), expression (1) can be written in another way:

$$\frac{\Delta \lambda}{\lambda_0} = \frac{\Delta n_h}{n_h} + \frac{\Delta d_h}{d_h} + B_2 \left[\left(\frac{\Delta n_l}{n_l} - \frac{\Delta n_h}{n_h} \right) + \left(\frac{\Delta d_l}{d_l} - \frac{\Delta d_h}{d_h} \right) \right]. \tag{4}$$

We may assume that the differences between the relative changes of the indices of refraction and those of the geometrical thicknesses of low- and high-index layers, are small and multiplied with the coefficient B_2 (B_2 is in our case of group B filters much less than unity) can be neglected with respect to the first two terms on the r.h.s. of relation (4). Thus relation (4) reduces to the approximative form:

$$\frac{\Delta \lambda}{\lambda_0} \approx \frac{\Delta n_{ZnS}}{n_{ZnS}} + \frac{\Delta d_{ZnS}}{d_{ZnS}} \tag{5}$$

where subscript h is replaced with subscript ZnS.

The relative change of the peak transmittance wavelength observed after neutron bombardment of the filters is in this way related both to the relative changes of the ZnS index of refraction and to the geometrical thickness of layers. The latter quantity can be calculated using the simple displacement theory2). Thus, the total number of atomic displacements, N_d, in the solid after irradiation with neutron flux Φ is expressed by

$$N_d = N_A \sigma_d \Phi \bar{\nu}. \tag{6}$$

Here N_A is the number of atoms per cm^3 of irradiated material, σ_d is the neutron cross section for the displacement and $\bar{\nu}$ is the mean number of atoms displaced by the primary. It has been estimated that the volume increase of irradiated material per each displaced atom (or each vacancy-interstitial pair) would amount to twice the atomic volume, though some calculations indicate that a value of 1.5 might be more appropriate[3]. Thus, knowing the number of displaced atoms per unit volume of irradiated material, one can calculate the relative volume increase, or the corresponding density decrease, $\frac{\Delta\varrho}{\varrho}$, which gives the relative change of geometrical thickness; namely, assuming that the relative dimensional changes in all three dimensions are the same, the following relation holds:

$$\frac{\Delta\varrho}{\varrho} = -3\frac{\Delta d}{d}.$$

Now let us calculate the number of displaced atoms in the ZnS layer of a filter belonging to group B, after a flux of $5 \cdot 10^{17}$ neutrons/cm^2 (see Fig. 2); in this case, the experimentally determined quantity $\frac{\Delta\lambda}{\lambda_0}$ is -0.00245, and we can, by means of relation (5), determine the relative change of the ZnS index of refraction. The mean number of atoms displaced by the primary, $\bar{\nu}$, for the case of reactor neutrons is given by the expression:

$$\bar{\nu} \approx \frac{E_p\,(\mathrm{Max})}{4E_d\left(\ln\frac{E_p\,(\mathrm{Max})}{E_d} - 1\right)}. \tag{7}$$

Here E_d is the threshold displacement energy usually taken to be 25 eV, and E_p (Max) is the maximum energy of the primary given by

$$E_p\,(\mathrm{Max}) \approx \frac{4E_n}{A} \qquad \text{if } A \gg 1, \tag{8}$$

where A is the atomic weight of atoms of irradiated material and E_n is the neutron energy. Taking into account the energy distribution of neutrons of TRIGA reactor, one obtains for the mean number of atoms displaced by the primary, $\bar{\nu}$, the value of 235. Since the neutron cross section σ_d for the displacement of Zn and S atoms is approximately $3 \cdot 10^{-24}$ cm^2, the number of displaced atoms in unit volume of ZnS layers after a flux of $5 \cdot 10^{17}$ neutrons/cm^2 is $N_d = 1.79 \cdot 10^{19}$ cm^{-3}. Assuming that the volume increase per each displaced atom is 1.5 times the atomic volume, the value obtained for N_d leads to $\frac{\Delta d_{ZnS}}{d_{ZnS}} = +0.000176$, so that the index of refraction of ZnS layers decreases by 0.26%. The decrease of index of refraction may be expected in both kinds of layers, because the introduction of a large number of displaced atoms and large disordered regions by energetic neutrons, leads to a »looser« structure which, in the case of high neutron fluxes, tends to become amorphous. X-ray diffraction analysis of filters has shown that ZnS layers are mostly polycrystalline before irradiation, where both a cubic, and to a smaller extent, a hexagonal phase were present. After bombardment, the peaks corresponding to the reflections from the planes of both the cubic phase and the hexagonal phase were broadened, indicating the onset of amorphization.

As it has already been shown, bombardment also induces broadening and splitting of the transmittance band of the filter into two bands. The broadening of the band is probably a consequence of the changes in optical thicknesses of the reflector layers; however, at the moment it is difficult to explain the splitting of the band which might be connected with the introduction of the amorphous phase in the layers. In any case, a detailed study of bombardment effects in cryolite and ZnS layers is necessary in order to elucidate the observed changes in optical properties of the filters.

References

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UTJECAJ NEUTRONSKOG OZRAČAVANJA NA SVOJSTVA FABRY-PEROTOVIH INTERFERENCIONIH FILTARA

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Ispitivan je utjecaj ozračavanja reaktorskim neutronima na optička svojstva Fabry-Perotovih interferencionih filtara iz ZnS i kriolita. Snimani su spektri optičke transmisije prije i poslije ozračavanja; korišteni su fluksevi neutrona od 10^{13}—10^{18} neutrona/cm^2. Kod neutronskih flukseva većih od 10^{17} neutrona/cm^2 opažen je pomak valne duljine maksimalne transmisije kao i smanjenje transmisije. Promjena valne duljine maksimalne transmisije povezana je s promjenom optičke debljine slojeva, tj. s promjenom indeksa loma i geometrijske debljine, koji su prouzrokovani neutronskim ozračavanjem. Izračunato je smanjenje gustoće i indeksa loma slojeva ZnS koristeći eksperimentalne rezultate i jednostavnu teoriju pomaka.