Solar Control on Irradiated Ta_2O_5 Thin Film

Nilgün DOĞAN BAYDOĞAN[a], Esra ÖZKAN ZAYİM[b] *

[a]Institute of Energy, [b]Science and Literature Faculty
Istanbul Technical University, Ayazaga Campus, Maslak, 34469, Istanbul, Turkey

Abstract
Ta_2O_5 thin films were prepared by sol-gel prosses on Corning 2947 glass substrates to obtain good quality films. These films were placed around the source and exposed to gamma radiation at 0.35, 2.58 and 4.5 kGy absorbed dose at room temperature using Co-60 radioisotope. Ta_2O_5 coated thin films were irradiated with beta radiation at 0.13 kGy using Sr-90 radioisotope. It is suggested that the mechanism of absorption is related to an allowed indirect transition at Ta_2O_5 film. When the absorbed dose of Ta_2O_5 films increased from 0.35 kGy to 4.5 kGy the optical band gaps of Ta_2O_5 films decreased from 2.40 eV to 1.63 eV 24 hours later from the irradiation proces using Co-60 radioisotope. But, the optical band gap of irradiated Ta_2O_5 film increased again 44 months later from the irradiation proces. The optical band gaps of irradiated Ta_2O_5 film at 0.35 kGy and 4.5 kGy are detected as 2.43 eV to 1.75 eV. The effect of gamma irradiation on the solar properties of Ta_2O_5 films is compared with that of beta irradiation. Using the optical properties, the redistribution of the absorbed component of the solar radiation and the shading coefficient (sc) are calculated as a function of the convective heat-transfer coefficient in order to achieve the control of solar energy. When the absorbed dose increased the shading coefficient decreased considerably. Changes in shading coefficient of irradiated Ta_2O_5 films are performed 24 hours later and 44 months later from the irradiation process in order to determine post irradiation behavior of Ta_2O_5 films. Shading coefficient of irradiated Ta_2O_5 films increased manifestly 44 months later from the irradiaiton. The shading coefficient of the irradiated Ta_2O_5 films with Sr-90 radioisotope differ significantly from irradiated Ta_2O_5 film with Co-60 radioisotope. Beta irradiation is more effective than gamma irradiation on the change of shading coefficient of Ta_2O_5 films.

Corresponding Author: Esra Özkan Zayim, Tel: +90.0212.285.30.09, ozesra@itu.edu.tr

Keywords: Co-60, Sr-90, Sol-gel, Irradiation, Solar properties, Ta_2O_5 Thin films

Introduction

Thin films consisting of Ta_2O_5 have been used in industrial applications related to thin-film capacitors, optical waveguides, and antireflection coatings on solar cells. Ta_2O_5 films are used for several special application areas as highly refractive material and show different optical properties depending on the deposition methods [1-6].

Coated glass materials can be used broadly depending on optical and solar performance of them. Under irradiation, the defect centers in these materials formed as a result of charge trapping by radiolytic electrons or holes. One of the essential processes occurring irradiation of the samples is the formation of electron–hole pairs. These free carriers can move and recombine, so that the photoelectrons are trapped at structural defects or impurities such as oxygen vacancies and multivalent impurities, while the holes are self-trapped at bridging or non-bridging oxygens. These new electronic configurations give rise to some preferential high absorption levels called color centers [7-10].

There are several types of radiation such as direct and/or indirect ionizing radiation and illumination radiation at high dose areas, irradiation constructions, the hot cells and the peeping holes for the specific researches. For the radiation damages with Co-60 radioisotope and Sr-90 radioisotope lead to variations in optical properties of Ta_2O_5 films. The mechanisim of optical transition is investigated in order to define the optical band gap. The changes on shading coefficient at Ta_2O_5 film on Corning 2947 are based on the absorbed radiation as cumulative dose in this study. This paper presents some data on solar control in terms of shading coefficient and its post irradiation behavior to obtain a complete study.

2. Solar Parameters

For the convenience of EN (European Norm) and CEN (European Committee for Standardization) standards to solar parameters in EN 410, the investigation of solar properties is important to know the shading coefficient in the solar range (300-2100 nm) including infrared terrestrial window region (800-1300 nm). The total solar energy transmittance, g, is calculated as the sum of the solar direct transmittance τ_e and the secondary heat transfer factor q_i of the glazing towards inside (Eq.1) according to EN (European Norm) at the solar range. Solar control is achieved by absorption only exhibits optimum effect if it is positioned externally. This becomes clear from the definition of the g value [11].

$$g = \tau_e + q_i \tag{1}$$

The effectiveness of solar control mechanism can be made clear using Eq.2 [11].

$$\tau_e + \rho_e + \alpha_e = 1 \tag{2}$$

The absorption term ($\alpha_e\phi_e$) is subsequently split into two parts $q_i\phi_e$ and $q_o\phi_e$ which are energy transferred to the inside and outside respectively (Eq.3);

$$\alpha_e = q_i + q_o \tag{3}$$

Where, q_i is the secondary heat transfer factor of the glazing towards the outside. The solar direct transmittance (τ_e) of the glazing is calculated using Eq.4.

$$\tau_e = \frac{\sum_{300}^{2500} S_\lambda \tau(\lambda)\Delta\lambda}{\sum_{300}^{2500} S_\lambda \Delta\lambda} \tag{4}$$

S_λ is relative spectral distribution of the solar radiation and $\tau(\lambda)$ is the spectral transmittance of the glazing. It is considered that radiation has the spectral composition of global radiation (direct sun at a height of 90^o and corresponding diffuse radiation from sky). Contrary to real situation at glass in building, it is always assumed, for simplification, that the spectral distribution of the solar radiation is not dependent upon atmospheric conditions (e.g. dust, mist, moisture content) and that the solar radiation strikes the glazing as a collimated beam and at normal incidence. The resulting errors are very small [13, 14].

The corresponding values $S_\lambda \Delta\lambda$ are given in such a way that $\Sigma S_\lambda \Delta\lambda = 1$. The solar direct reflectance (ρ_e) of the glazing is calculated using Eq.5. $\rho(\lambda)$ is the spectral reflectance of the glazing [13, 14].

$$\rho_e = \frac{\sum_{300}^{2500} S_\lambda \rho(\lambda) \Delta\lambda}{\sum_{300}^{2500} S_\lambda \Delta\lambda} \qquad \textbf{(5)}$$

For the calculation of the secondary heat transfer factor towards the inside q_i, coefficients of the glazing towards the outside h_e and towards the inside h_i, are needed. These values mainly depend on the position of the glazing, wind velocity, inside and outside temperatures and furthermore on the temperature of the two internal glazing surfaces. The purpose of this standard is to provide basic information on the performance of glazing. The conventional conditions have been stated for simplicity: Position of glazing: vertical, Outside surface: wind velocity (~ 4m/s), corrected emmisivity=0.837, Inside surface: natural convection, emissivity optional, air space are unventilated, under these conventional average conditions, standard values for h_e and h_i are obtained: h_e= 23 W/(m^{2}K), h_i=8 W/(m^{2}K).

Secondary internal heat transfer factor q_i, of a single glazing calculated using the Eq.6 [13, 14].

$$q_i = \alpha_e h_i / (h_e + h_i) \qquad (6)$$

The shading coefficient is a ratio of the ability of the clear glass to reject solar heat gain [13-14]. This coefficient of a glazing is the ratio between the quantity penetrating through the glazing and the energy that was penetrating into it through a single glass, in the same irradiation of direct radiation alone is concerned. It is related with the solar heat gain that the solar energy transmitted through a glazing plus the portion of solar radiation that is absorbed and either convected or reradiated inwards [15]. The shading coefficient calculated dividing the solar factor of the glazing in question by that of the clear glass. The value of it standardized by the American Society of Heating, Refrigerating and Air Conditioning Engineers (ASHRAE) in the U. S. A. is 0.88. It enables air-conditioning engineers to adapt all the tables and formula for single clear glass to the case of special glazing [17].

3. Experimental Conditions

Soda lime glass gives the good results at obtaining adherent film. Sodium, which diffuses out of soda-lime glass, supports the film and acts as a surfactant at the film [18]. Corning 2947, microscope slide substrate is accepted as a standard soda-lime glass [19]. Therefore the usage of Corning 2947 known as standard soda-lime-silicate glass is a suitable substrate with 1 mm thickness for evaluation of optical and solar changes according to declared standards. The optical and solar properties of clear soda-lime-silica glass are standardized with 1 mm thickness at the solar range by CEN in 1992 [14, 17].

3.1. Experimental Details on Sol-Gel Process

The optical and structural properties of the thin films could be tailored easily by sol-gel deposition methods [20]. Porous films with a large surface area that lead to improved kinetic performance are desirable [21-23]. Hence it is thought the use of sol-gel procedure for the preparing of film has some advantages for the investigation of solar parameters at this study.

Ta_2O_5 films were deposited by a spin-coating process. Tantalum oxide coating solutions were prepared by hydrolysis of tantalum ethoxide in absolute ethanol. The tantalum ethoxide was dissolved in ethanol and the molar ratio was 0.25. After the solution had been stirred for 30 min, it was diluted with ethanol and a small amount of water mixture. The mixed solution was catalyzed with glacial acetic acid and stirred for 18 h. The obtained solution was transparent. The spin coating speed was set at 2000 rpm. The thickness of Ta_2O_5 films is about 150 nm.

3.2. Irradiation Process

The Co-60 and Sr-90 radioisotopes is an appropriate source to change the color and to create colour centers Ta_2O_5 films on Corning 2947 substrate as this substrate can be accept as a soda-lime silica [7].

Cumulative dose is the total dose resulting from repeated exposures of ionizing radiation to the same portion over a period of time. When a material is used in either terrestrial or space applications, it is exposed to radiation fields that are uniform or non-uniform. Radiation fields may vary with geometry or with time [12]. Therefore cumulative dose of the materials is the important parameter as the absorbed dose of material at the place of interest during their missions. So, test samples were exposed to irradiation at 0.35, 2.58 and 4.50 kGy using Co-60 radioisotope and at 0.13 kGy using Sr-90 radioisotope at the room temperature.

After irradiaiton processes with Co-60 and Sr-90 radioisotopes the colour of all samples tuned to brownish colour. Irradiated samples were kept a dark place at the room temperature for 44 months in order to reduce the light and temperature effect at the variations of the induced color centers to permissible levels before measuring the optical properties again. The colour of irradiated samples bleached considerably 44 months later.

4. Results and Discussion

Soda-lime-silicate glass is a type of the glass, which consists of SiO_2 70%, Na_2O 15% mainly. The addition of soda (Na_2O) to silica lowers the softening point by 800-900^{o}C. Alumina (Al_2O_3) are added to improve chemical resistance in soda-lime silicate glass. This glass is used especially sheet (including windows), containers and light bulbs [15]. The transmittance and absorption of soda-lime-silica glass depends on the purity and state of oxidation (stoichiometry) of silica [11]. Ionic impurities and colour centers as the transition elements are important to know the reason of colour. The detection of transition elements at Ta_2O_5 film on Corning 2947 was made using Innov-X XRF Analyzer and results given in Table 1**.**

The transmittance and reflectance of all samples at the range between 280 - 2500 nm was measured using a spectrophotometer Perkin Elmer Lambda 9 UV/VIS/NIS. The results of transmittance and absorbance were given in Fig.1. The irradiated Ta_2O_5 films deposited by spin coating technique are non-absorbing beyond ~ 300 nm and the absorption edges are similar. The optical transmittance of irradiated Ta_2O_5 films varies considerably in the visible region depending on absorbed dose and when the post irradiaiton time increases transmittance of the irradiated samples increases.

For indirect transition materials $\alpha(E)$ is given by $\alpha(E) = A(E - E_g)^2$ where $\alpha = 4\pi k/\lambda$ is absorption coefficient, E = hv is the photon energy, and E_g is the band gap, respectively. E_g for allowed indirect transition was determined by extrapolating the linear part of $(\alpha E)^{1/2}$ vs. E plot, shown in Fig.2, to α=0 (E is the energy of light in eV). The optical band gap of irradiated sample at 0.35 kGy increased from 2.40 to 2.43 eV due to the new electronic distribution when the post irradiation time reached from 24 hours to 44 months respectively. When the absorbed dose increased to 4.5 kGy, the optical band gap decreased to 1.63 eV and when post irradiation time reached 44 months optical band gap was 1.75 eV. The increase of absorbed dose and elapsed time from the irradiation process are very important to explain the changes of optical band gap due to the interaction of the electromagnetic wave with the electron in the valance band. If Ta_2O_5 film on Corning 2947 glass is exposed to high level absorbed dose such as 0.35 kGy, the generated radiation defects seems to be meta-stable depending on post irradiation time.

Ionic impurities, colour centres and defects depending on the purity and state of oxidation (stoichiometry) are important to know that optical density is the reason for the change of colour of Ta_2O_5 film on glass material. Evaluation of optical density was made using Eq.7 [8]. There are D, optical density; t, film thickness; α, absorbance and ρ, reflectance in Eq.7. The evaluation of the reflectance together with absorbance is important for the examination of the color centers.

$$D=\alpha t \log_{10} e-\log_{10}(1-\rho)^2 \qquad (7)$$

When the absorbed dose of the samples increased, a characteristic optical density band occurred at ~ 630 nm in Fig.3. The changes in optical density indicate that the coloration is a result of the creation of induced defect centers at this band. The induced optical density band at ~ 630 nm is sited 44 months later from the irradiation process. However there is a decrease at the optical density depending on the elapsed time after the irradiation process.

For the evaluation of the effectiveness of solar control mechanism of the test samples, the changes of calculated solar direct transmittance (τ_e), reflectance (ρ_e), and absorbance (α_e) via the absorbed dose were shown in Fig. 4. There are not so much changes of solar direct reflectance when the absorbed dose and post irradiation time increases. Direct solar absorbance, (α_e) of the samples increases while direct transmittance, (τ_e) decreases as a function of absorbed dose. However, there are the increase of (τ_e) and decrease of (α_e) when post irradiation time increases. Effect of post irradiation time on solar direct transmittance and absorbance is observed when the absorbed dose increases.

For the description of the transferred proportion of absorbed solar energy, secondary heat transfer factor of inside q_i and outside q_o was shown in Fig.5. When the absorbed dose increases secondary heat transfer factor rises. Effect of post irradiation time on secondary heat transfer factors can be detected easily when the absorbed dose increases.

Shading coefficient of induced Ta_2O_5 film on Corning 2947 decreased when the absorbed dose increased in Fig.6. Shading ability enhanced depending on an increase of total solar heat transmittance and the increase of the absorbed dose improved the shading coefficient. However the shading coefficient changes depending on elapsed time after the irradiation process. The shading coefficient increased sharply 44 months later from the irradiation process.

Besides, the effect of beta irradiation on shading coefficient is compared with that of gamma irradiation in Fig.6. The energy of electrons from Sr-90 source (by yttrium emission) is high enough to produce colour centers. 0.546 MeV is the maximum electron energy emitted by Sr-90. However there is the yttrium emission and its electron is more energetic (~ 2.2 MeV). The shading coefficient of the irradiated sample at 0.13 kGy with Sr-90 is sited on the shading coefficient curve obtained from the irradiated samples with Co-60. It can be thought the induced color centers at 0.13 kGy with Sr-90 affected the shading coefficient considerably. The effect of irradiation with Sr-90 seemed more powerful than the effect of irradiation with Co-60 on change of shading coefficient.

5. Conclusions

The transition elements are responsible to the enhancement of color by the increase of optical density. Gamma and beta irradiation generated a characteristic optical band at ~ 630 nm. The created band has addressed to the transition elements such as Zr, and Re from Corning 2947 glass substrate and Ta from Ta_2O_5 film. Besides, the changes of the optical density bands due to absorbed dose explained the causes of the enhanced coloration by induced color centers.

Induced Ta_2O_5 film on Corning 2947 by Co-60 reduces the incidence of solar radiation considerably. Shading coefficient decreased sharply using Co-60 when the absorbed dose increased. When the absorbed dose increased from 0.35 kGy to 4.5 kGy shading coefficient was reduced ~ 13.5 % 24 hours later from the irradiation process. However, the shading coefficient started to increase depending on the increase of elapsed time after the irradiation process. The reduction of shading coefficient is ~ 10.5 % 44 months later from the irradiation process. Besides, the electrons from Sr-90 source may provide adequate penetration at Ta_2O_5 film and interact with the sample effectively. Improved shading coefficient by beta irradiation can enable effective solar energy saving at the hot climate or special research areas.

Changes at shading coefficient as the amount of heat passing through inside have related with the increase of induced colour centers at both Ta_2O_5 film and the glass structure. Created color centers have changed the performance of shading coefficient considerably and improved shading coefficient has addressed the noteworthy gain in solar energy at Ta_2O_5 film on glass. The best performance may be obtained from an irradiated Ta_2O_5 film on glass with a lower shading coefficient at a hot climate, special study conditions or experimental media.

REFERENCES

[1]F. Tepehan et al., Determination of optical properties of amorphous Ta_2O_5 films deposited by spin- and dip-coating methods, Solar Energy Materials and Solar Cells 46, (1997), 311-321.

[2]F. E. Ghodsi and F. Tepehan, Heat treatment e!ects on the optical properties of Sol-gel Ta_2O_5 thin films, Solar Energy Materials & Solar Cells 59, (1999), 367-375.

[3]F. Tepehan et al., Optical properties of sol-gel dip-coated Ta_2O_5 films for electrochromic applications, Solar Energy Materials & Solar Cells, 59, (1999) 265-275.

[4]J. M. Ngaruiya et al., Preparation and characterization of tantalum oxide films produced by reactive DC magnetron sputtering, phys. stat. sol. (a) 198, No. 1, 99– 110 (2003)

[5]Cheng-Chung Leea and Der-Jun Jana, Optical properties and deposition rate of sputtered Ta2O5 films deposited by ion-beam oxidation, Thin Solid Films 483 (2005) 130– 135

[6]S. Boughabaa et al., Synthesis of tantalum pentoxide Ælms by pulsed laser deposition: material characterization and scale-up, Thin Solid Films 358 (2000) 104±113

[7]Baydogan, N.D., Tugrul, A.B., Glass Physics and Chemistry 32 (3), pp. 309-314 (2006), Evaluation of the Optical Changes for a Soda–Lime–Silicate Glass Exposed to Radiation

[8]Zayim, E.O., Baydogan, N.D., Solar Energy Materials and Solar Cells 90 (4), pp. 402-413 (2006)

[9]ESR dosimeter based on P2O5–CaO–Na2O glass system, Z.M. Da Costa, W.M. Pontuschka, J.M. Giehl, C.R. Da Costa, Journal of Non-Crystalline Solids 352 (2006) 3663–3667

[10] N. Dogan, A. B. Tugrul, Proceedings of 15th European TRIGA Conference, VTT Symposium 197, Espoo-Finland, (1997) 81-102.

[11] Dogan, N., Tugrul, AB, Optical and solar parameters of irradiated lead-alkali-silicate glass, Solar Energy Materials & Solar Cells 69 (2001) 241-250.

[12] Bouquet, F., Spacecraft design-thermal and radiation, an overview of thermophysics and nuclear radiation design technology, (Systems Co., Inc., Carsborg, Washington, 1991).

[13] N. Dogan, A. B. Tugrul, Radiation Measurements, 33-2, (2001) 211-216.

[14] EN 410, European Standard, (1998), Glass in building-Determination of luminous and solar characteristics of glazing, ICS 81.040.20, CEN-European Committee for Standardization, Ref.No. EN 410:1998 E.

[15] CEN-European Committee for Standardization, (1992), Glass in Builging-Determination of Light transmittance, solar direct transmittance, total solar energy transmittance and ultraviolet transmittance, and related glazing characteristics, CEN/TC 129/WG9/N.4E Rew.7, (Final Draft prEN 410).

[16] J. S. Amstock, Handbook of Glass in Conctruction, (McGraw-Hill, New-York 1997).

[17] H., J., Glaser, Large Area Coating, (Von Ardenne Anlagen Technk GMBH, Dresden, 2000)

[18] CEC - Commission of the European Communities, Reported by European Group of Flat Glass Producers (GEPVP), Industrial Process Building and Civil Engineering, Flat Glass for Building, (1983) Brussels, Belgium, EUR 8069 DE, EN, FR, IT, Directorate-General for the Internal Market and Industrial Affairs.

[19] K. Ellmer, C. Stock, K. Diesner, I. Sieber, Journal of Crystal Growth, 182 (1997) 389-393.

[20] C. L. Hansen, E. Skordalakes, J. M. Berger, S. R. Quake, Applied Physical Sciences PNAS-Proceedings of the National Academy of Sciences of the United States of America, 99-26 (2002) 16531–16536

[21] E. Ozkan and F. Z. Tepehan, Solar Energy Materials and Solar Cells, 68, Issues 3-4, (2001), 265-277

[22] E. Ozkan Zayim, Ping Liu, Se-Hee Lee, C. E. Tracy, J. A. Turner, J. R. Pitts and S. K. Deb, Solid State Ionics, 165, 1-4, (2003), 65-72.

[23] E. Ozkan, Se-Hee Lee, C. E. Tracy, J. R. Pitts and S. K. Deb, Solar Energy Materials and Solar Cells, 79, 4, (2003), 439-448

TABLE CAPTIONS

Table-1. The results of XRF Analysis of transition elements at Ta_2O_5 film on Corning 2947.

FIGURE CAPTIONS

Fig.1. Transmittance T (%) and reflectance R (%) of Ta_2O_5 films on Corning 2947.

Fig.2. Allowed indirect transition and optical band gap of Ta_2O_5 films on Corning 2947.

Fig.3. Optical density of Ta_2O_5 films on Corning 2947 at doses of (1) 0.35 kGy by Co-60, (2) 2.58 kGy by Co-60, (3) 0.13 kGy by Sr-90, (4) 4.5 kGy by Co-60.

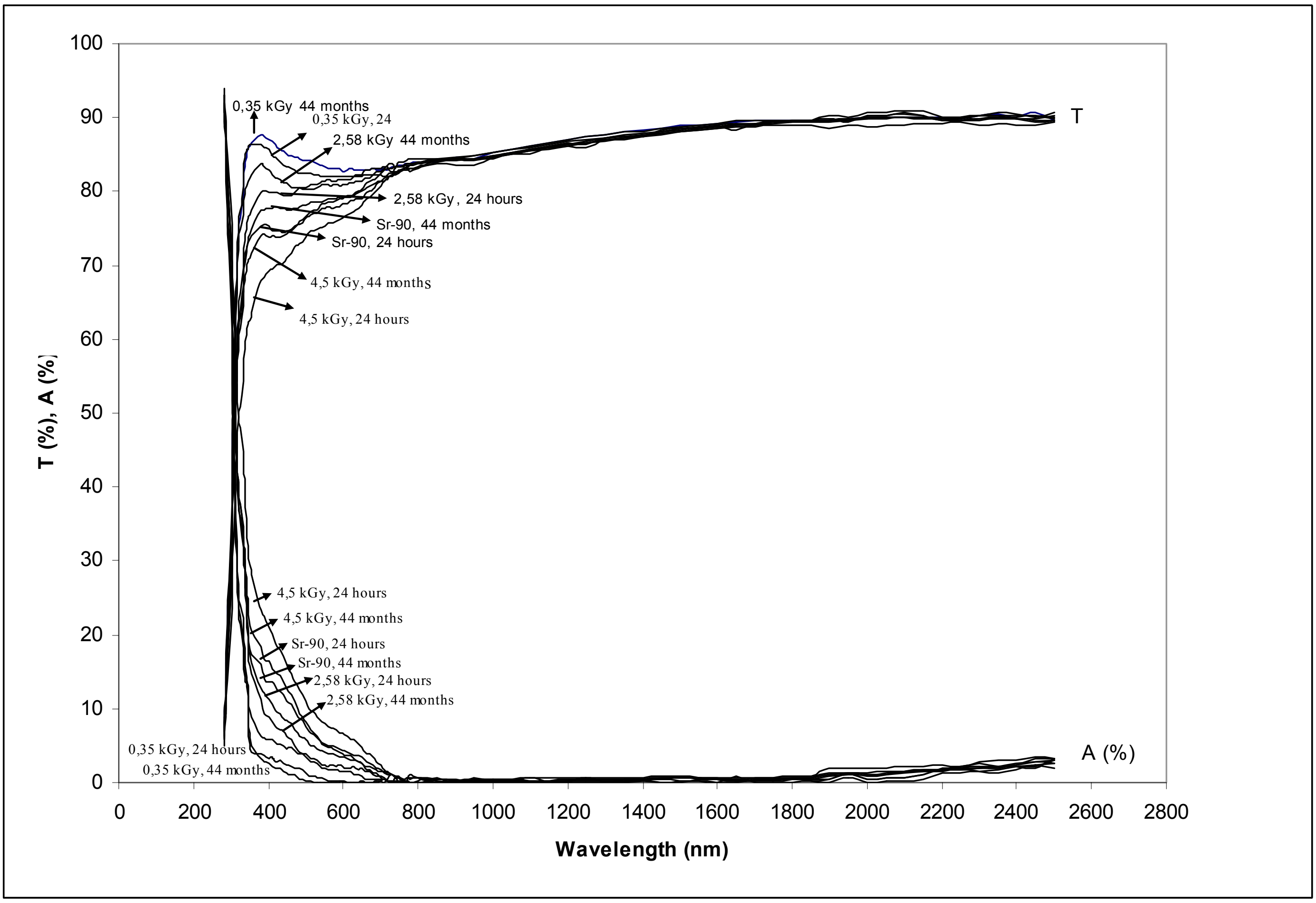
Fig.4. Solar Direct Transmittance τ_e (%), Reflectance ρ_e (%) and Absorbance α_e (%) of Ta_2O_5 films on Corning 2947depending on absorbed dose.

Fig.5. Secondary heat transfer factors of Ta_2O_5 films on Corning 2947.

Fig.6. Shading coefficient of Ta_2O_5 films on Corning 2947.

Table-1.

Elements	%	+/-
Ta	0,13	0,02
Re	0,03	0,00
Zr	0,01	0,00



100
90
80
70
60
50
40
30
20
10
0
T (%), A (%)
0,35 kGy 44 months
0,35 kGy, 24
2,58 kGy 44 months
2,58 kGy, 24 hours
Sr-90, 44 months
Sr-90, 24 hours
4,5 kGy, 44 months
4,5 kGy, 24 hours
T
4,5 kGy, 24 hours
4,5 kGy, 44 months
Sr-90, 24 hours
Sr-90, 44 months
2,58 kGy, 24 hours
2,58 kGy, 44 months
0,35 kGy, 24 hours
0,35 kGy, 44 months
A (%)
0 200 400 600 800 1000 1200 1400 1600 1800 2000 2200 2400 2600 2800
Wavelength (nm)

Fig.1

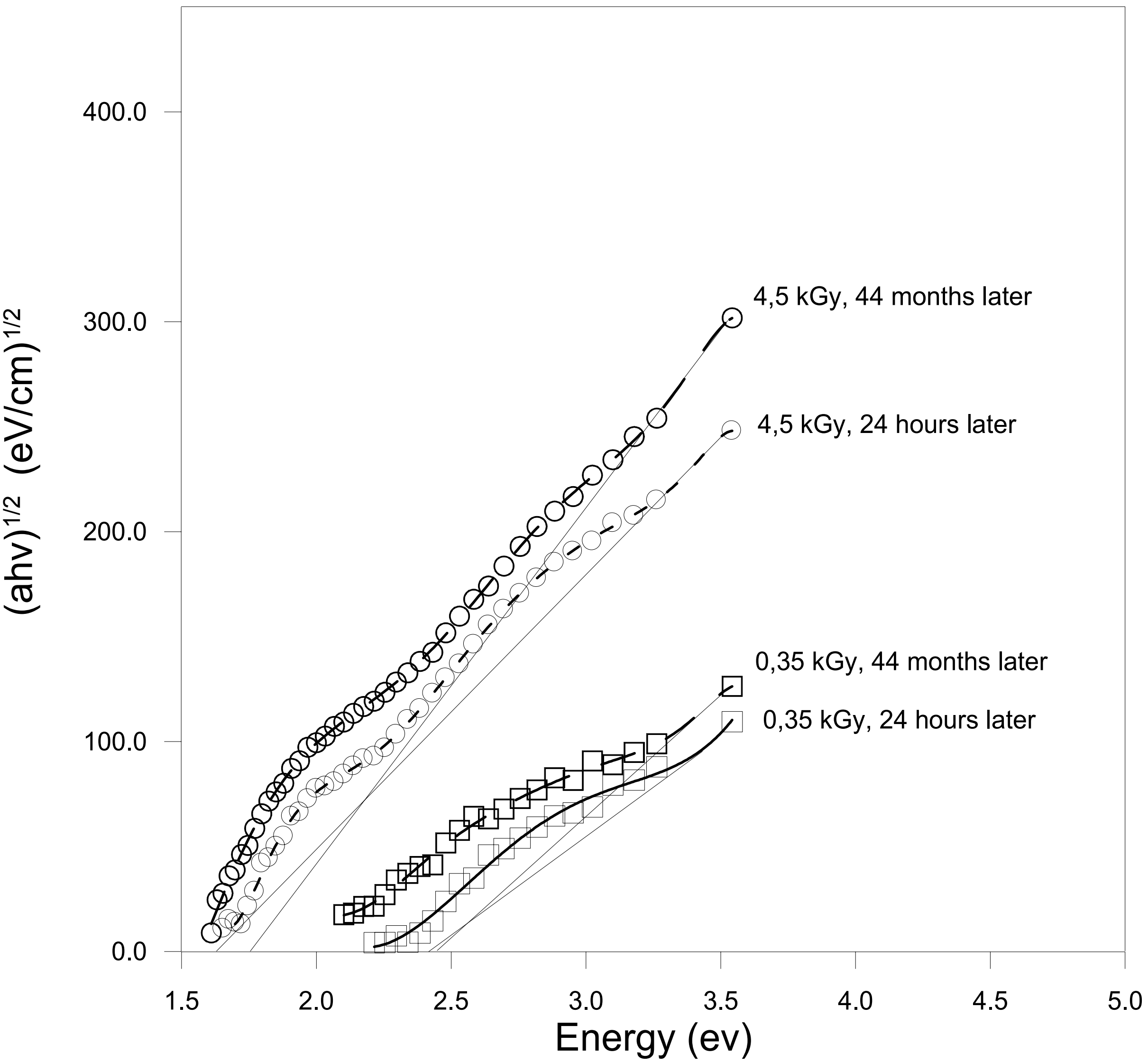
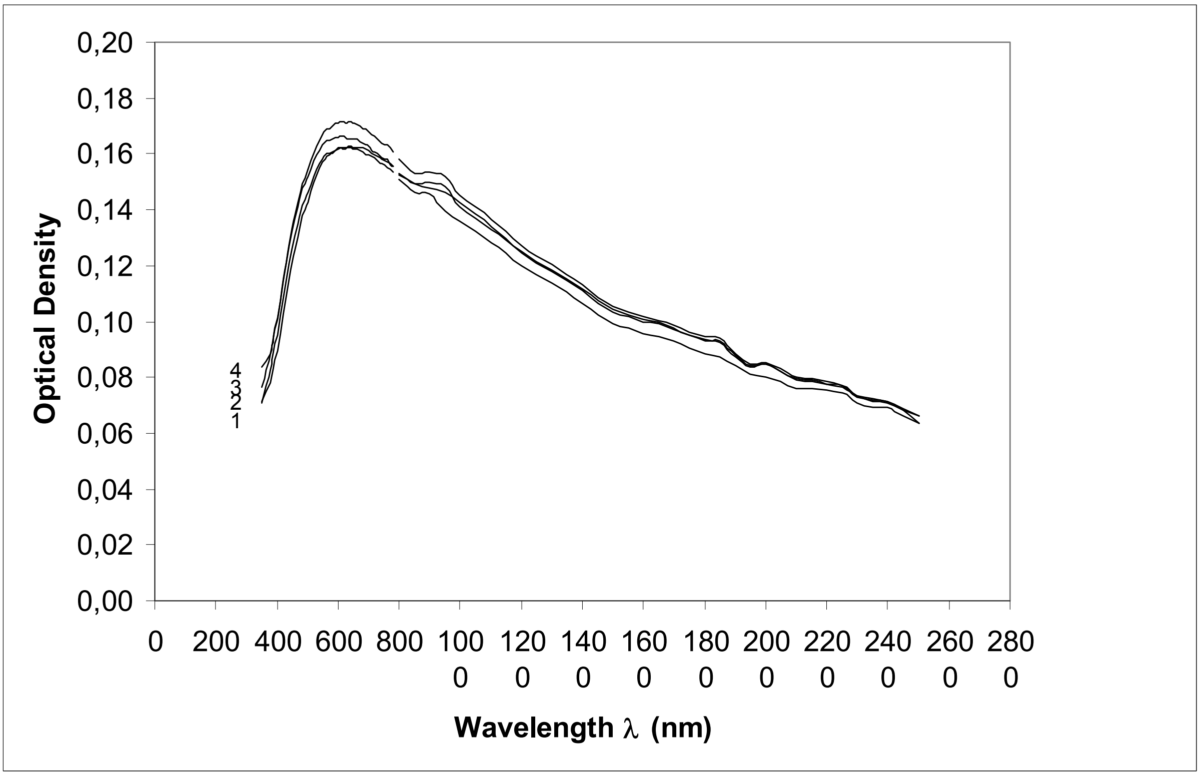
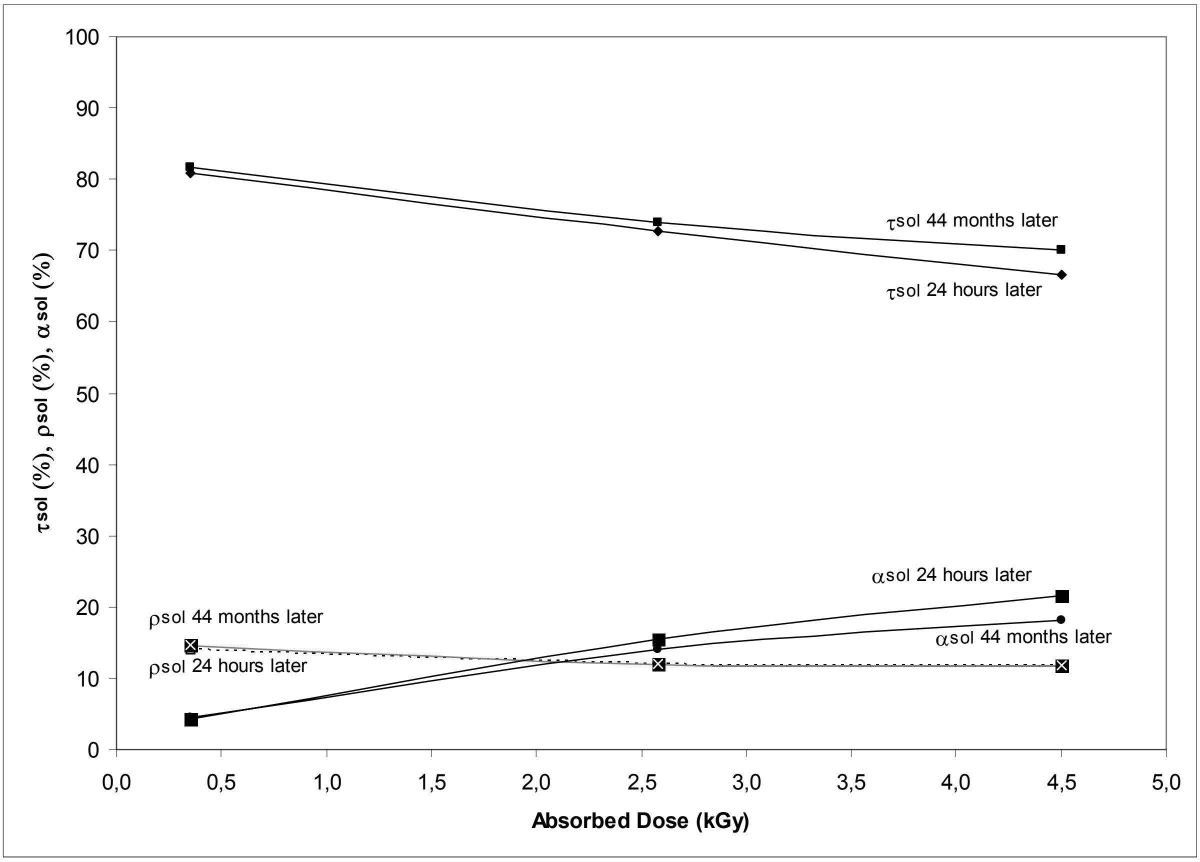

4,5 kGy, 44 months later
4,5 kGy, 24 hours later
0,35 kGy, 44 months later
0,35 kGy, 24 hours later
(ahv)^1/2 (eV/cm)^1/2
Energy (ev)
0.0
100.0
200.0
300.0
400.0
1.5
2.0
2.5
3.0
3.5
4.0
4.5
5.0

Fig.2



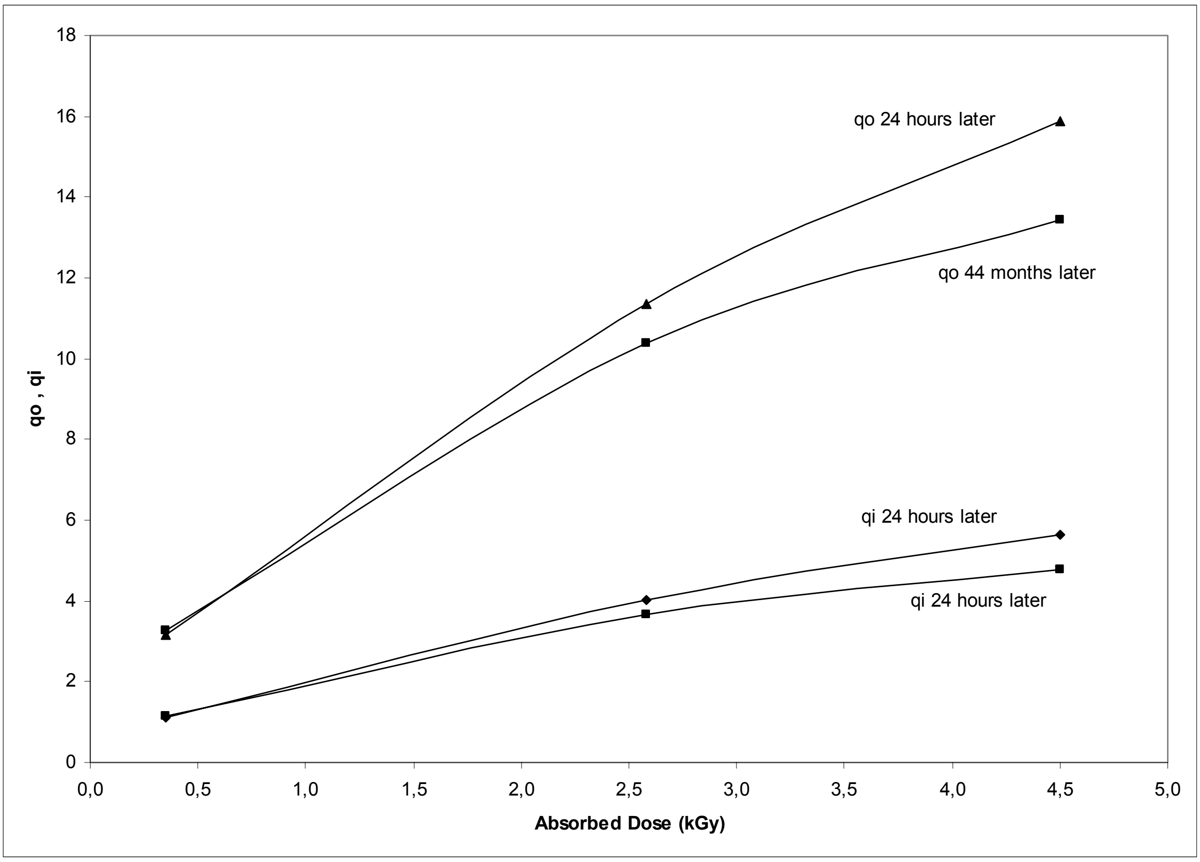
0,20
0,18
0,16
0,14
0,12
0,10
0,08
0,06
0,04
0,02
0,00
Optical Density
4
3
2
1
0 200 400 600 800 100 0 120 0 140 0 160 0 180 0 200 0 220 0 240 0 260 0 280 0
Wavelength λ (nm)

Fig.3



100
90
80
70
60
50
40
30
20
10
0
τsol (%), ρsol (%), αsol (%)
τsol 44 months later
τsol 24 hours later
αsol 24 hours later
ρsol 44 months later
αsol 44 months later
ρsol 24 hours later
0,0
0,5
1,0
1,5
2,0
2,5
3,0
3,5
4,0
4,5
5,0
Absorbed Dose (kGy)

Fig.4



qo 24 hours later
qo 44 months later
qi 24 hours later
qi 24 hours later
qo , qi
Absorbed Dose (kGy)
0
2
4
6
8
10
12
14
16
18
0,0
0,5
1,0
1,5
2,0
2,5
3,0
3,5
4,0
4,5
5,0

Fig.5

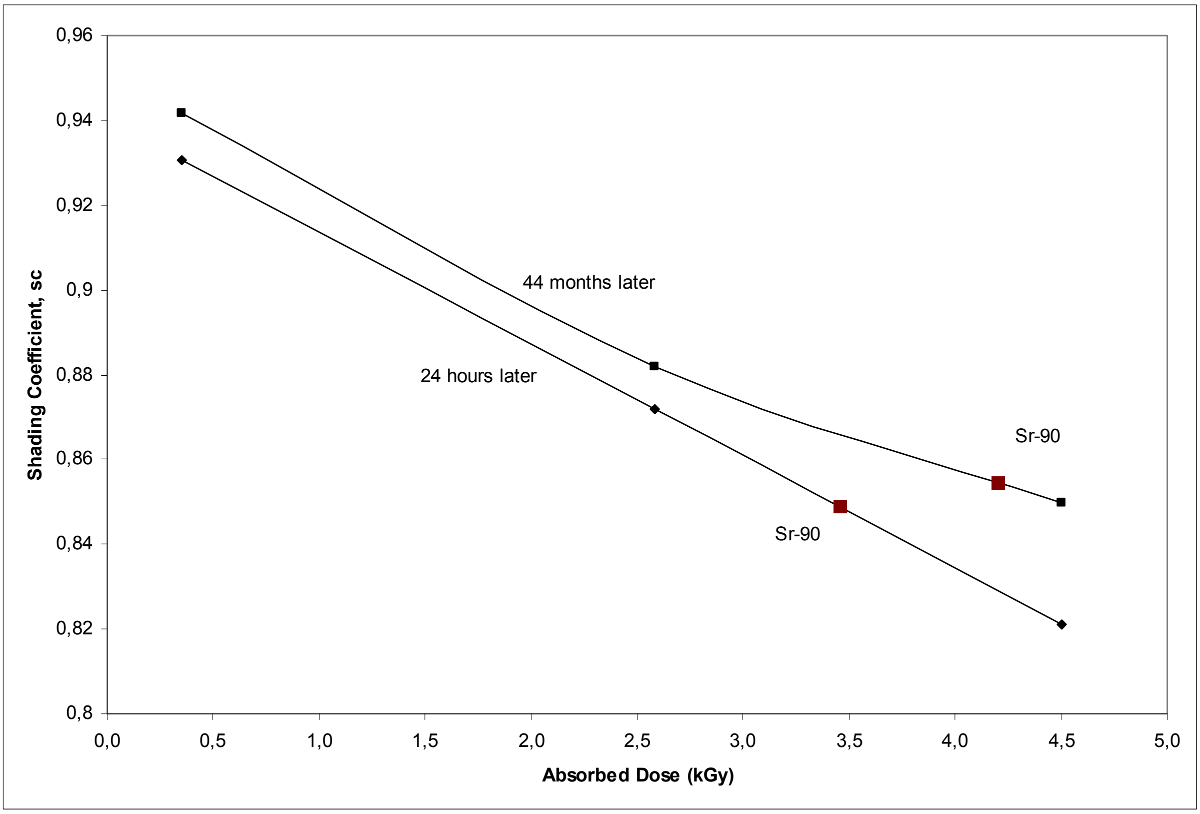

0,96
0,94
0,92
0,9
0,88
0,86
0,84
0,82
0,8
Shading Coefficient, sc
0,0
0,5
1,0
1,5
2,0
2,5
3,0
3,5
4,0
4,5
5,0
Absorbed Dose (kGy)
44 months later
24 hours later
Sr-90
Sr-90

Fig.6