

## AGING INFLUENCE ON THE THERMAL AND OPTICAL PROPERTIES OF ARTISTIC GLASSES: EXPERIMENTAL DATA AND COMPARISON

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### ABSTRACT

The aim of the present paper is to evaluate the influence of the aging process on the thermal and optical properties of glasses employed in the restoration of artistic windows. Therefore, 10 glass samples of different color, thickness and staining technique were examined. Half of each sample was subject to an accelerated aging process of 11 weeks in a climatic chamber (see Fig. A-1). Then, in order to evaluate the influence of the thermal stress and strain due to environmental conditions such as air temperature and relative humidity on the behavior of the glasses, the thermal expansion coefficient  $\alpha$  of the new and aged samples was measured. Measurements were carried out in the same climatic chamber used for the aging process (see Fig. A-1), employing suitable strain gauges. Results show that the aging process could produce significant differences between the values obtained for the new and the aged samples. A general trend of increased  $\alpha$  for the aged samples was noticed.

The influence of the aging process on the radiative properties of the same samples was investigated also.

Specifically, the spectral transmission and reflection coefficients were measured with a spectrophotometer Cary 2300 (see Fig. A-2); the light transmittance and reflectance and the solar direct transmittance and reflectance were calculated. Results show that the aging process influences only the reflection properties in the visible wavelength range of sample No. 6, the only one stained with a particular technique called as “grisaglia” (in the box of Fig. A2); the reflection coefficient of the aged sample is greater than that of the new one; the light reflectance of the aged sample is twice that of the new one. Results suggested the extended research to 5 other samples, all stained with the grisaglia technique; four of them are antique fragments from the Arezzo and Cortona Cathedrals, one of them is a new sample employed in the restoration of the artistic windows. Results show that the antique samples have higher values of the reflection coefficients. The reason is that the aging process makes the glass surfaces decorated with the grisaglia technique smoother.



Fig. A-1: The climatic chamber employed for the aging process and the thermal expansion coefficient measurements.



Fig. A-2: The spectrophotometer Cary 2300 employed for the optical properties measurements; in the box sample No. 6 is illustrated.

## INTRODUCTION

The artistic glasses decorating churches and cathedrals play an important role in the sacred representation and also in daylight.

Umbria Region has many artistic windows in its churches: the big one in the Saint Domenico's Church in Perugia is only an example. So the conservation of these art works is very important. The environmental conditions (temperature, relative humidity, solar radiation) could damage the artistic glasses, due to the thermal stress and strain generated by the surface temperature excursions; during centuries the glasses have been subject to very different environmental conditions, so the aging process on their surface could be significant.

In the present paper, the influence of the aging process on the thermal and optical properties of glasses used in the restoration of artistic windows is evaluated; so a group of ten different samples were first examined: half of each sample was subject to an accelerated aging process in climatic chamber. Then the thermal expansion coefficient of the new and aged samples was measured, in the same climatic chamber, employing strain gauges glued on the glass sample surface; finally a comparison between the results obtained for the new and the aged samples was carried out.

The optical properties of the same samples were also investigated; in particular the spectral transmission and reflection coefficients were measured in the range of 180-3150 nm, employing a spectrophotometer Cary 2300. The light transmittance and reflectance and the solar direct transmittance and reflectance were also calculated; the values related to new and aged samples were finally compared. The aging process influence was found in a sample stained with a particular technique called *grisaglia*. So the research was extended to a second group of 5 samples all stained with this technique: 4 of them are antique fragments from the Cathedral of Arezzo and Cortona windows, one is a new glass employed for restoration. The same optical measurements and calculations were repeated and a comparison between the new and the antique samples was carried out.

## NOMENCLATURE

$r$  = spectral reflection coefficient (-).

$s$  = thickness (mm);

$t$  = spectral transmission coefficient (-);

$T$  = temperature ( $^{\circ}\text{C}$ , K);

$T_{max}$  = maximum temperature in measurement cycle ( $^{\circ}\text{C}$ );

$T_{min}$  = minimum temperature in measurement cycle ( $^{\circ}\text{C}$ ).

### Greek Letters

$\alpha$  = coefficient of linear thermal expansion ( $^{\circ}\text{C}^{-1}$ ,  $\text{K}^{-1}$ );

$\Delta\varepsilon$  = strain gauge signal (variation between sample and reference material) (-);

$\Delta\varepsilon_{T_{max}}$  = strain gauge signal at  $T = T_{max}$ ;

$\Delta\varepsilon_{T_{min}}$  = strain gauge signal at  $T = T_{min}$ ;

$\lambda$  = wavelength (nm);

$\rho_v$  = light reflectance (-);

$\rho_e$  = solar direct reflectance (-);

$\tau_v$  = light transmittance (-);

$\tau_e$  = solar direct transmittance (-);

$\phi$  = relative humidity (%).

### Subscripts

$A$  = aged sample;

$N$  = new sample;

$R$  = reference bar;

$S$  = sample.

## CHARACTERISTICS OF THE SAMPLES

Two groups of samples were examined: the first group was composed of all the different kinds of glasses employed in the artistic windows restoration: different colors, different thicknesses, different staining techniques; the second group was composed of different samples stained with the *grisaglia* technique. Only the samples of the first group were subject to the aging process; each sample was cut into two parts and only one of these parts of each sample was aged while the other one was maintained new. The major steps of the *grisaglia* technique is that the sheet of glass is painted employing a mixture of fine ground iron and copper oxides, bound by glass powder and borax; then it is put inside a kiln at  $600^{\circ}\text{C}$ : the mixture, vitrifying during the natural cooling, sticks to the glass support. This technique is used not only to stain the glass, but to decorate it also (see Fig. 1).

### Preliminary samples

The first group of samples includes ten glasses; samples No. 1, 2, 3, 4, 7, 8, 9, 10 are of different colors and thickness and are all stained in the glass paste; the coloration of sample No. 5, red, is obtained in a stained bath; sample No. 6, rose-pink (flesh colored), is stained with the *grisaglia* technique (see Table 1).

### Grisaglia samples

The second group of samples includes five glasses of different colors, all stained with the *grisaglia* technique; samples No. 11, 12 and 13 are small fragments of the antique Arezzo Cathedral glass; sample No. 14 is a small piece of an ancient glass in Cortona; sample No. 15 is a modern *grisaglia* decorated glass representing the Virgin Mary and the Baby Jesus (Fig. 1).



Fig. 1: sample No. 15, decorated with the *grisaglia* technique.

## AGING PROCESS

The aging process was realized in compliance with UNI 10593-2 [1], referring to insulating glasses. It consists of two parts:

- 56 cycles of 12 hours, composed of a ramp between  $T = -18^{\circ}\text{C}$  and  $T = 53^{\circ}\text{C}$  with a temperature gradient of  $14^{\circ}\text{C/h}$  (5 hours), a maintenance period at  $T = 53^{\circ}\text{C}$  (1 hour), a descending ramp between  $T = 53^{\circ}\text{C}$  and  $T = -18^{\circ}\text{C}$  with a temperature gradient of  $-14^{\circ}\text{C/h}$  (5 hours) and a maintenance period at  $T = -18^{\circ}\text{C}$  (1 hour); the relative humidity is always greater than 95% and the whole period is 4 weeks;
- a period of 7 weeks with  $T = 58^{\circ}\text{C}$  and  $\Phi$  greater than 95%.

The whole period of the aging process is 11 weeks. It was realized employing a Mazzali climatic chamber, the same employed for the thermal expansion coefficient measurements, from March 27th 2001 to June 12th 2001. During the process the surface temperature of the glasses was measured.

Table 1: Characteristics of the first group of samples.

Sample	Color	Coloration Technique	Notes	s (mm)
1	violet	glass paste	ovoidal bubbles	2.71
2	colorless	glass paste	ovoidal bubbles	2.71
3	violet	glass paste	scrapes on both x and y sides	3.74
4	green	glass paste	spherical bubbles	2.84
5	red	bath	bath on both x and y sides	2.65
6	rose-pink	grisaglia	grisaglia only on the x side	1.96
7	blue	glass paste	scrapes on both x and y sides	3.96
8	blue	glass paste	very small bubbles	3.39
9	yellow	glass paste	very small bubbles	3.52
10	violet	glass paste	small spherical bubbles	2.38

## THERMAL EXPANSION COEFFICIENT MEASUREMENTS

The thermal expansion coefficient was measured in order to evaluate the thermal stress and strain produced on the artistic windows by environmental conditions such as temperature and solar radiation; if too high, these could produce a fragile rupture of the glass. This behavior could be emphasized by the natural aging process.

### Experimental facility and procedure

The experimental facility was described in a previous paper [2], in which the results of the thermal expansion coefficient of the glasses of the first group before the aging process were reported. The facility is constituted by a Mazzali climatic chamber mod. C33045 in which the samples are placed. The thermal expansion coefficient  $\alpha$  was measured employing strain gauges glued over the samples; twin strain gauges were chosen, placed one over the sample, one over a reference material (silica-titanium bar), with  $\alpha_R = 0.03 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ , in order to eliminate the error due to the thermal expansion of the strain gauge grid. The strain gauge signal yields the value

of the strain variation between sample and reference material  $\Delta\epsilon$  and is recorded by a multichannel transducer SPIDER 8 connected to a PC for the data storage and analysis. At the same time, the surface temperature of the samples and of the silica-titanium bar are measured with NTC sensors (Negative Temperature Coefficient): a tarature curve was evaluated to transform the electrical voltage in the corresponding temperature value.

The experimental procedure, such as described in [2], is composed of three phases:

- stability of the strain gauge signal evaluation, in order to verify, at  $T = \text{cost} = 24^{\circ}\text{C}$ , that the strain gauge signal is about zero; such a verification is periodically carried out during all the measurements;
- residual stresses elimination in all the components of the measurements system (glass samples, reference bar, strain gauges, wires, etc.), by means of 3 cycles of temperature at least  $5^{\circ}\text{C}$  above the maximum value and below the minimal value of the measurement cycle; each cycle is composed of a ramp between  $T = 5^{\circ}\text{C}$  and  $T = 55^{\circ}\text{C}$ , with temperature gradient of  $12.5 \text{ }^{\circ}\text{C/h}$ , a maintenance period of 40 min. at  $T = 55^{\circ}\text{C}$ , a descending ramp between  $T = 55^{\circ}\text{C}$  and  $T = 5^{\circ}\text{C}$  with a temperature gradient of  $-12.5^{\circ}\text{C/h}$  and a final maintenance period of 40 min. at  $T = 5^{\circ}\text{C}$ ; the total time of the three cycles is 28 hours;
- measurement cycle, composed of a maintenance at  $T = 10^{\circ}\text{C}$  for 4 hours, a ramp with a temperature gradient of  $6.66^{\circ}\text{C/h}$  until a value of  $T = 50^{\circ}\text{C}$  (6 hours), a maintenance at  $T = 50^{\circ}\text{C}$  for 4 hours, a descending ramp with a temperature gradient of  $-6.66 \text{ }^{\circ}\text{C/h}$  (6 hours) and a final maintenance at  $T = 10^{\circ}\text{C}$  for 4 hours; during the cycle the values of  $\Delta\epsilon$  and  $T$  are recorded, in order to evaluate the thermal expansion coefficient  $\alpha$ , given by [2]:

$$\alpha_s = \alpha_R + \frac{\Delta\epsilon_{T_{\max}} - \Delta\epsilon_{T_{\min}}}{T_{\max} - T_{\min}} \quad (1)$$

## Results

The thermal expansion coefficient  $\alpha$  was measured for the 10 samples of the first group; 20 values of  $\alpha$  were measured, 10 before and 10 after the aging process, in order to verify the influence of the aging process on the thermal expansion properties; the samples before the aging process are called NEW (N), the samples after the aging process are called AGED (A).

For each sample at least 3 tests were carried out, in order to verify the possibility of repeating the results; as an example, in Fig. 2 the results related to test II on aged sample No. 9 are shown. The surface temperature trend vs. time and the relative strain  $\Delta\epsilon$  were measured; the corresponding value of  $\alpha$  was evaluated employing relation (1). Fig. 2 shows the strain trend vs. increasing and decreasing temperature: there is not a hysteresis loop, so the hypothesis of uniformity throughout the thickness of the sample is verified. For the sake of brevity, only the

calculated values of  $\alpha$  of the other samples are reported in Table 2.

Measurements related to the new samples, reported in [2], were repeated measuring the strain of the new and aged samples contemporarily. A comparison between the thermal expansion coefficient of the new and aged samples is shown in Fig. 3.

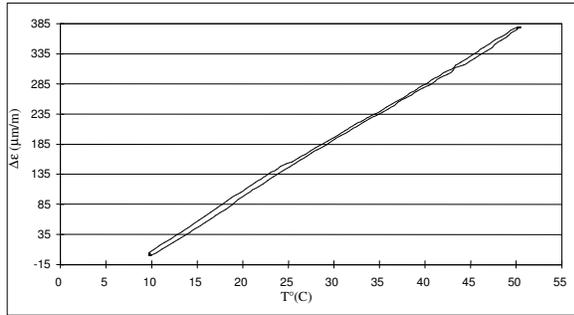


Fig. 2: Strain vs. temperature during the test (aged sample No. 9).

Table 2: Thermal expansion coefficient of the 10 samples new (N) and aged (A) and comparison.

Sample	s (mm)	$\alpha$ ( $\mu\text{m}/\text{m}^\circ\text{C}$ )		
		N	A	A-N
1	2.71	8.63	9.38	+0.75
2	2.71	8.78	10.44	+1.66
3	3.74	8.35	9.57	+1.22
4	2.84	9.74	9.76	+0.02
5	2.65	9.54	10.82	+1.28
6	1.96	9.62	9.58	-0.04
7	3.96	8.24	8.85	+0.61
8	3.39	8.08	8.71	+0.63
9	3.52	9.22	9.61	+0.39
10	2.38	8.91	10.66	+1.75

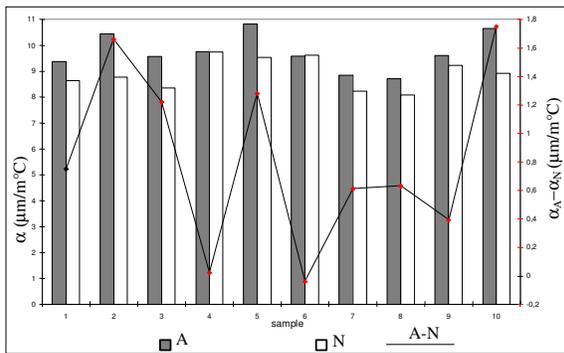


Fig. 3: Comparison between the thermal expansion coefficients of the new and aged samples.

Except for sample No. 6 ( $\alpha_A - \alpha_N = -0.04 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ ), where the difference is so small to be comprised in experimental uncertainty, the difference ( $\alpha_A - \alpha_N$ ) is always positive, so the aging process increases the thermal expansion coefficient.

The difference ( $\alpha_A - \alpha_N$ ) varies in the range  $0 - 1,75 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ , so it is about  $0 - 20\%$  of the new corresponding sample. The samples with a more evident influence of the aging process are No. 2, 5 and 10 ( $+1,28 - 1,75 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$ ); these samples are characterized by a slight thickness. Only sample No. 6 has a slighter thickness, but it is the only one stained with the grisaglia technique and it does not present any influence of the aging process on the thermal expansion coefficient. A statistical test of significance (t-test) was performed in the Table 2 data. The probability value that the data are from the same population is very low and equal to  $0.25\%$ ; so the two series of data (new and aged values of  $\alpha$ ) could belong to the same population (but the data are representative of a rare event) or they belong to different populations. In this case the aging process influence could be important.

In order to evaluate if the material is isotropic, some tests with three-axial strain gauges were carried out. As an example, results related to the aged sample No. 8 are shown in Fig. 4: the strain values in the three directions are approximately the same, so it can be concluded that the material is isotropic and mono-axial strain gauges can be used.

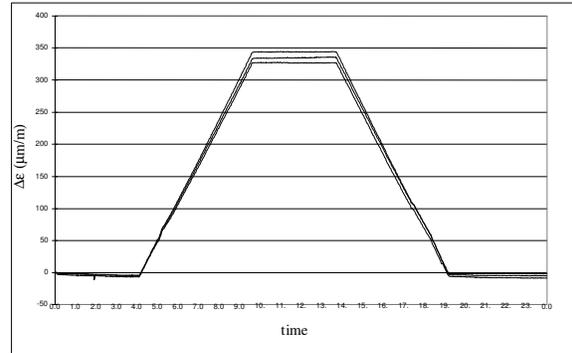


Fig. 4: Three – axial strain vs. time during the isotropic test (aged sample No. 8).

#### Measurement uncertainty

The measurement uncertainty has been evaluated in [2], in compliance with UNI CEV ENV 13005 [3]; the  $\alpha$  measurement uncertainty varies between  $0,17$  and  $0,21 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$  for the different samples: it is about  $2\%$  of the measured value.

#### TRANSMISSION AND REFLECTION COEFFICIENT MEASUREMENTS

The transmission and reflection coefficients were measured for the samples of the first group; then, because of the influence of the aging process on the optical properties of grisaglia stained sample No. 6, research was extended to the samples of the second group.

#### Experimental facility and procedure

The spectral transmission and reflection coefficients were measured employing a spectrophotometer Cary 2300; the spectral range of wavelength is  $\lambda = 185 - 3152 \text{ nm}$ . The spectrophotometer, in the standard configuration,

allows to measure the spectral transmission coefficient of low diffusive material; in the configuration including the integrating sphere, it allows to determine the spectral reflection coefficient and the spectral transmission coefficient of diffusive materials. A detailed description of the instrumentation is reported in [4].

The sample preparation and the measurement procedure were carried out in compliance with UNI 7885 [5] and EN 410 [6]. The spectral transmission coefficient measurements were carried out in the standard configuration and were repeated with the integrating sphere, in order to evaluate possible diffusive behavior of the sample.

The spectral reflection coefficient was measured for both sides of each sample, called side x and side y.

Finally the calculations of the light transmittance and light reflectance (wavelength range of 380-780 nm) and of the solar direct transmittance and solar direct reflectance (wavelength of 300-2500 nm) was carried out, in compliance with EN 410 [6].

Sample No. 15 is too big to be introduced in the spectrophotometer Cary 2300; so, for the measurements on this sample, a portable spectrophotometer S2000 was employed; the spectral range wavelength is 300-1015 nm and it is equipped with optical fibers for the transmission and reflection measurements. In the transmission measurements two optical fibers are employed; one of them is fed by a radiation source and supplies the signal, while the other one measures the radiation signal; the sample is placed between the two fibers and the ratio between the two signals gives the transmission coefficient. In the reflection measurements only one optical fiber is employed, different from the ones employed for the transmission measurements; it supplies the radiation signal and, in the mean time, measures the radiation reflected by the sample: the ratio between the two signals gives the reflection coefficient.

For sample No. 15 the spectral transmission and reflection coefficients were measured in different spots and only the light transmittance and reflectance were evaluated, in compliance with EN 410 [6].

## Results

The detailed results related to the samples of the first group were reported in [4]; in particular:

- the samples stained in the glass paste (1, 2, 3, 4, 7, 8, 9, 10) do not have diffusive behavior, so the transmission results obtained in the standard configuration of the spectrophotometer and with the integrating sphere are coincident;
- samples No. 5 (stained bath) e No. 6 (stained with the grisaglia technique) have diffusive behavior, so only the transmission measurements carried out with the integrating sphere are reliable;
- the aging process does not influence the spectral transmission coefficient (as an example in Fig. 5 the results related to sample No. 7 new and aged are reported);
- the spectral reflection coefficient is the same for the

two sides x and y of each sample new and aged, except for sample No. 6;

- for all the samples, except for No. 6, the aging process does not influence the spectral reflection coefficient;
- sample No. 6 has a spectral reflection coefficient higher on side x, stained with the grisaglia technique, for both the new and the aged sample;
- sides x of the new and aged samples No. 6 have different spectral reflection coefficient: the aged sample has a higher value of  $r(\lambda)$  (Fig. 6); so the aging process influences the spectral reflection coefficient in the visible wavelength range.

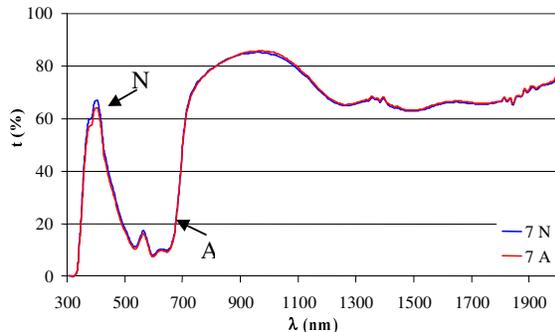


Fig. 5: Spectral transmission coefficients of the new (N) and aged (A) sample No. 7

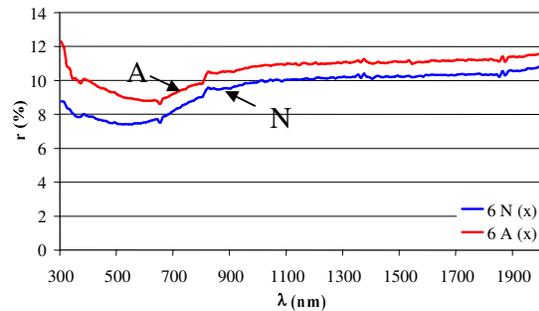


Fig. 6: Spectral reflection coefficients of the new (N) and aged (A) sample No. 6 (sides x).

The values of the light transmittance  $\tau_v$  and reflectance  $\rho_v$  and of the solar direct transmittance and reflectance  $\rho_e$  and  $\rho_e$  of the ten new and aged samples, calculated in compliance with EN 410 [6], are reported in Table 3: the aging process does not influence these parameters, except for the light transmittance on side x of sample No. 6, painted with the grisaglia technique; in this case the  $\rho_v$  value of the aged sample is about twice the value of the new one ( $\rho_{vN}=0,0479$ ;  $\rho_{vA}=0,0895$ ).

Results obtained for the reflection properties of sample No. 6 encouraged to study in detail the grisaglia painted glasses; so the spectrophotometer measurements on the second group of samples (No. 11, 12, 13, 14, 15) were carried out. The spectral transmission and reflection coefficients of samples No. 11, 12, 13 and 14 are reported in Fig. 7 (a, b, c, d): the antique glasses have transmission coefficients variable in the range 5-80%, while the reflection coefficients are always lower than 15%.

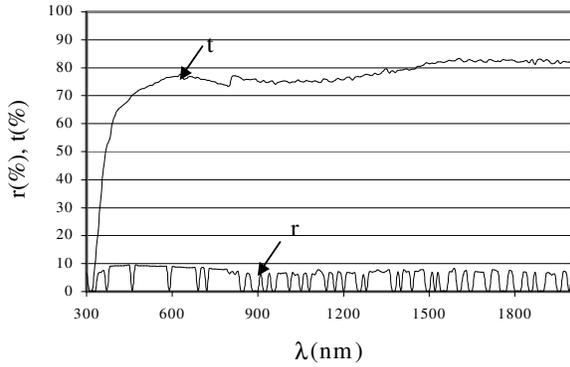


Fig. 7a: spectral transmission and reflection coefficients for sample No. 11.

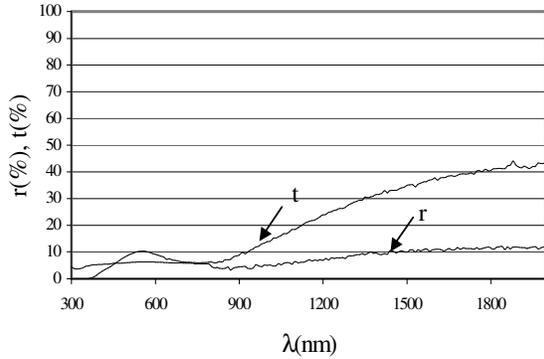


Fig. 7b: spectral transmission and reflection coefficients for sample No. 12.

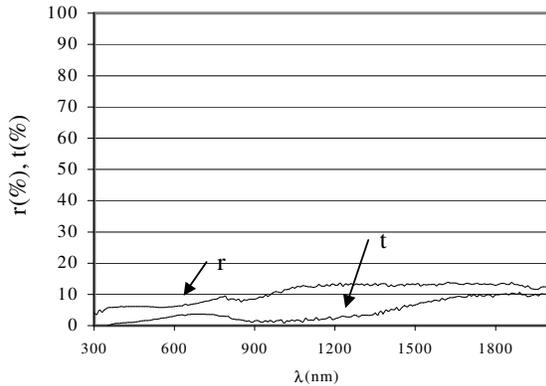


Fig. 7c: spectral transmission and reflection coefficients for sample No. 13.

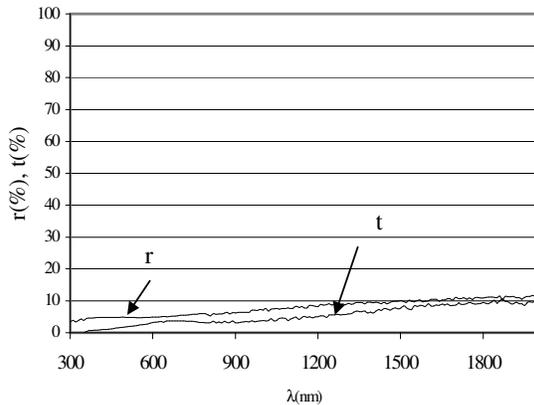


Fig. 7d: spectral transmission and reflection coefficients for sample No. 14.

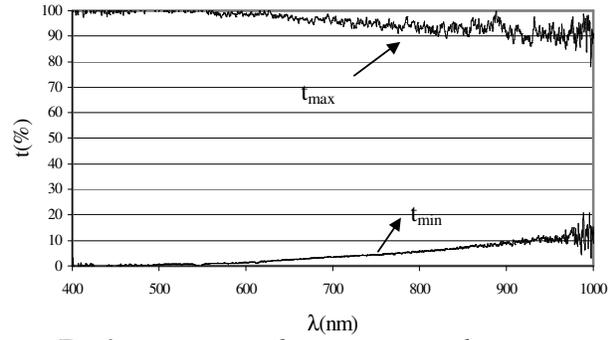


Fig. 8a: maximum and minimum spectral transmission coefficient for sample No. 15.

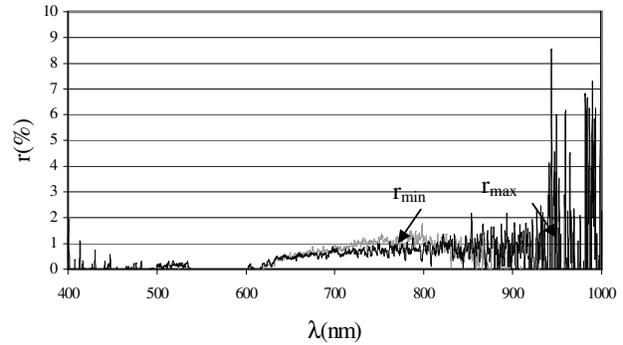


Fig. 8b: maximum and minimum reflection coefficient for sample No. 15.

Table 3:  $\tau_v$ ,  $\rho_v$ ,  $\tau_e$ ,  $\rho_e$  values for the new and aged samples of the first group; sample No. 6 has the maximum difference of  $\rho_v$  between the new and the aged sample.

Sample	Color	$\tau_v$	$\rho_v$	$\tau_e$	$\rho_e$
1	N Violet	0,0015	0,0466	0,2432	0,0493
	A (paste)	0,0017	0,0445	0,2548	0,0499
2	N Colorless	0,9159	0,0801	0,9021	0,0747
	A	0,8954	0,0809	0,8889	0,0756
3	N Violet	0,0096	0,0450	0,1784	0,0472
	A (paste)	0,0074	0,0471	0,1657	0,0491
4	N Green	0,0861	0,0456	0,3184	0,0523
	A (paste)	0,1224	0,0495	0,3419	0,0522
5	N Red	0,1445	0,0511	0,4788	0,0629
	A (bath)	0,1311	0,0508	0,4683	0,0608
6	N Pink	0,2121	0,0479	0,4556	0,0861
	A (grisaglia)	0,2287	0,0895	0,4764	0,0986
7	N Blu	0,1345	0,0499	0,4901	0,0632
	A (paste)	0,1254	0,0504	0,4837	0,0648
8	N Blue	0,0277	0,0440	0,4292	0,0570
	A (paste)	0,0260	0,0442	0,4084	0,0572
9	N Yellow	0,3913	0,0584	0,4111	0,0614
	A (paste)	0,3604	0,0585	0,4056	0,0631
10	N Violet	0,0168	0,0428	0,3626	0,0512
	A (paste)	0,0279	0,0433	0,3775	0,0532

For sample No. 15 the measurements were carried out in 15 different points; as an example, in Fig. 8 (a, b) the minimum and maximum spectral transmission and reflection coefficients are reported: the maximum value of the spectral transmission coefficient is in the range 90-99%, the minimum value is in the range 0-15%; the maximum and minimum values of the spectral reflection coefficient are approximately coincident and comprised

in the range 0-10%.

The calculation of  $\tau_v$ ,  $\tau_e$ ,  $\rho_v$  and  $\rho_e$  (see Table 4) shows that:

- the sides x of the antique samples No. 11, 12, 13 and 14, all decorated with the grisaglia technique, have values of  $\rho_v$  equal or higher than the maximum value of the new sample No. 15 (see Table 3);
- sample No. 13, with a double grisaglia sheet on the x side, has the lowest value of  $\rho_v$ : it shows a lower influence of the aging due to the time, because of the protective role of the second sheet;
- for sample No. 15,  $\tau_v$  varies in the range  $0,009 \div 0,933$ ;  $\rho_v$  varies in the range  $0,0001 \div 0,068$ ;
- the side y of sample No. 14, stained in color bath, has the higher value of  $\rho_v$ .

So it may be concluded that the antique samples have higher values of the reflection properties.

Table 4:  $\tau_v$ ,  $\tau_e$ ,  $\rho_v$  and  $\rho_e$  values for the samples of the second group.

Sample	$\tau_v$	$\tau_e$	$\rho_v$		$\rho_e$	
			x	y	x	y
11	0,750	0,710	0,084	0,082	0,056	0,069
12	0,094	0,110	0,062	0,064	0,059	0,068
13	0,024	0,031	0,058	0,048	0,062	0,058
14	0,024	0,026	0,060	0,083	0,081	0,079
15	min	0,009	-	$10^{-4}$	-	-
	max	0,933	-	0,068	-	-

### Uncertainty

The measurement uncertainty for the spectrophotometer Cary 2300 was evaluated in [7]; it is about 3% of the measured value for both transmission and reflection measurements.

### CONCLUSIONS

The environmental conditions (temperature, relative humidity) could produce in the artistic glasses degradation processes, due over all to their quick variations. In particular they could generate in glasses of different color and absorption coefficient different stress and strain conditions; it could be very dangerous for these materials characterized by fragile behavior. In very long periods, such as centuries, the environmental conditions could influence the transmission and reflection properties related to solar and visible radiation; this influence could modify daylight.

In the present paper the influence of an accelerated aging process on the properties of glasses employed in the restoration of artistic windows was carried out.

Ten samples of glass of different color, thickness and staining technique were firstly examined; each of them was divided into two parts: one was subject to an accelerated aging process in climatic chamber, the other one remained unaltered. The thermal expansion coefficients of the new and aged samples were measured employing strain gauge appropriate for thermal measurements. The comparison between the results obtained on the new and the aged samples shows that the aging process seems to influence the thermal expansion

coefficient values: the aged samples have  $\alpha$  values higher than the new samples; the difference  $\alpha_A - \alpha_N$  varies in the range  $0 - 1,75 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$  (about + 0 - 20% of the new corresponding sample). A statistical test of significance (t-test) showed that the two series of data ( $\alpha_A$  and  $\alpha_N$ ) could not belong to the same population of data, so the aging process could influence the thermal expansion coefficient of the glass samples.

The spectral transmission and reflection coefficients of the same samples were measured employing a spectrophotometer. The light transmittance and reflectance and the solar direct transmittance and reflectance were finally calculated. Results show that only the sample stained with a particular technique called grisaglia presents an influence of the aging process on its reflection properties; the aged sample No. 6, in fact, in the visible wavelength, has higher values of the reflection coefficient; the light reflectance of the aged sample is twice the light reflectance of the new one. So research has been extended to a group of 5 samples all stained with the grisaglia technique; results show that the antique samples from the Arezzo and Cortona Cathedrals have reflection properties higher than the new sample No. 15. So it may be concluded that the aging process makes the surface stained with the grisaglia technique smoother and it produces an improvement in the reflection properties.

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