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Study of ageing effects in aerogel

T. Bellunato^a, M. Calvi^a, C. Coluzza^b, G. Longo^b, C. Matteuzzi^a, M. Musy^a,
P. Negri^a, D.L. Perego^{a,*}

^a *Università degli Studi di Milano Bicocca and INFN, Milano 20126, Italy*

^b *INFN and Università di Roma “La Sapienza”, Roma, Italy*

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Abstract

Ageing effects on aerogel due to irradiation and absorption of humidity have been investigated. Aerogel tiles have been exposed to γ radiation from a ^{60}Co source and to proton and neutron high intensity beams. The transmittance has been monitored in the wavelength range between 200 and 800 nm, determining the clarity factor C as a function of the increasing dose of irradiation. The index of refraction n was also measured.

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1. Introduction

Silica aerogel is a solid material made of SiO_2 [1–3] with a very low density ranging between about 0.003 and 0.35 g/cm³. It is transparent and its refractive index can be tuned within the wide range of 1.008–1.08. Depending on the manufacturing procedure, the final product can be hygroscopic or hydrophobic. The use of aerogel as radiator for RICH detectors [4] demands extremely good optical quality [5–7]. This has now been achieved [8], and several experiments have built Ring Imaging Cherenkov detectors (RICH) with aerogel, like HERMES [9] or plan to build one, like LHCb [10], AMS [11] and BELLE [12].

Possible ageing of aerogel due to intense irradiation from different sources or absorption of humidity has been studied. Aerogel tiles were irradiated with very intense γ , proton and neutron beams or exposed to humid air. Optical parameters like transmittance, clarity and index of refraction were measured to monitor ageing. The hygroscopic aerogel used has been produced by the Borskov Institute of Catalysis in Novosibirsk [2]. The tiles used in the tests presented in this paper have dimensions of about 50 × 50 × 20 mm³ and the index of refraction n is around 1.03.

2. Optical properties of aerogel

The light diffusion within the aerogel is usually the factor limiting the performance of this material

*Corresponding author.

E-mail address: davide.perego@mib.infn.it (D.L. Perego).

URL: <http://castore.mib.infn.it/~dperego/>.

as Cherenkov radiator for a RICH detector. The dominant contribution to the total diffusion probability comes from the Rayleigh scattering mechanism. The scattering cross-section for a photon of wavelength λ is proportional to λ^{-4} .

The absorption cross-section is, within a wide range, wavelength independent and can be neglected when considering samples a few centimetres thick. The transmittance T of the aerogel can be parametrized by the Hunt formula [13]:

$$T(\lambda) = \frac{I}{I_0} = A e^{-Ct/\lambda^4}, \quad (1)$$

where I_0 and I are the incident and the transmitted intensities of the light beam through the sample. A is the surface scattering coefficient, t is the thickness of the aerogel block, and λ is the wavelength of the impinging light beam. C is the clarity factor and it is used to specify the optical quality of a sample together with A . An ideal aerogel would have A and C close to 1 and 0 respectively. Ageing effects have been monitored measuring the transmittance T and extracting the parameters A and C at several intermediate steps during irradiation and the exposure to humidity.

2.1. Transmittance measurements

The transmittance T was measured by means of a double beam spectrophotometer. A scan in the wavelength range between 200 and 800 nm was performed in steps of 1 nm.

An example of a measured spectrum is shown in Fig. 1 for a thickness of 21 mm. The curve is fitted with the Hunt formula (1) in order to determine the parameters A and C . The correlation coefficient between these two parameters is about 0.7.

To estimate the point to point variation of the Hunt parameters A and C , several measurements have been performed with the spectrophotometer beam entrance point scanning the two large ($50 \times 50 \text{ mm}^2$) surfaces. The dispersion observed in these measurements was assumed to be a systematic error on A and C for the whole tile. As shown in Fig. 2(a), for hygroscopic aerogel the transmittance at 400 nm is uniform in the tile at the level of 3–4%. The dispersion of the fitted value of the clarity C is about 2%, as shown in Fig. 2(b). In the

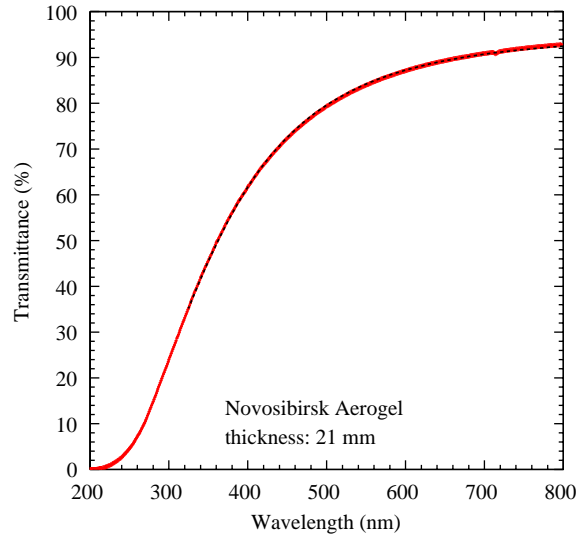


Fig. 1. An example of a transmittance measurement fitted with the Hunt relation (1). The result of the fit, shown as the black dotted line, is hardly visible because superimposed to the measurement. It gives $A = (94.97 \pm 0.04)\%$ and $C = (0.00526 \pm 0.00001) \mu\text{m}^4/\text{cm}$.

case of hydrophobic aerogel, these dispersions are about 1–2% for the transmittance at 400 nm and 7% for the clarity C respectively.

2.2. Measuring the index of refraction

The refractive index n of the aerogel was measured using green ($\lambda = 543.5 \text{ nm}$) and red ($\lambda = 632.8 \text{ nm}$) He–Ne laser sources. The measurements were performed using the prism method. Fig. 3 shows the block positioned in front of the laser beam on a rotating table. The sample is rotated until the deflection angle θ_{out} reaches its minimum. In that condition the index of refraction is given by:

$$n = \sin\left(\frac{\Phi + \theta_{\text{out}}}{2}\right) / \sin\left(\frac{\Phi}{2}\right), \quad (2)$$

where Φ is the angle between the two adjacent sides of the block ($\Phi \simeq 90^\circ$); θ_{out} , the deflection angle, is determined by measuring the displacement x with a CCD camera¹ located at $L = 870 \text{ mm}$ from the aerogel.

¹ SONY® XC-ST70CE with 768×512 pixels and a sensitive area coverage of $1.025 \times 0.850 \text{ cm}^2$.

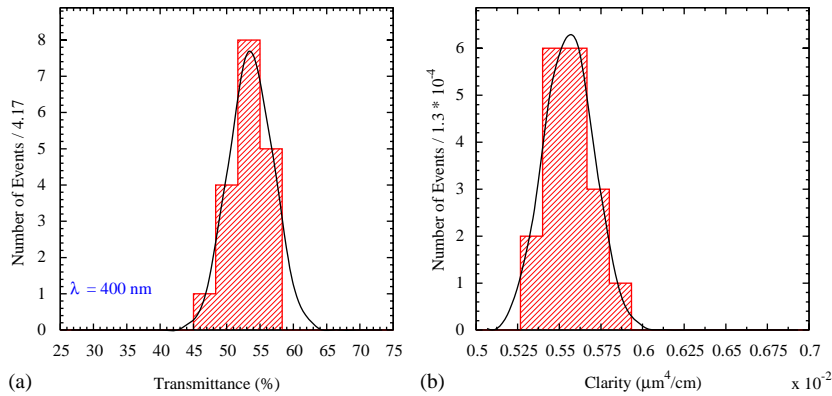


Fig. 2. Distributions for (a) transmittance T at $\lambda = 400$ nm and (b) clarity factor C . A Gaussian fit is superimposed, yielding $\sigma = 3.3\%$ for the transmittance at 400 nm and $\sigma = 0.13 \times 10^{-3} \mu\text{m}^4/\text{cm}$ for the clarity factor.

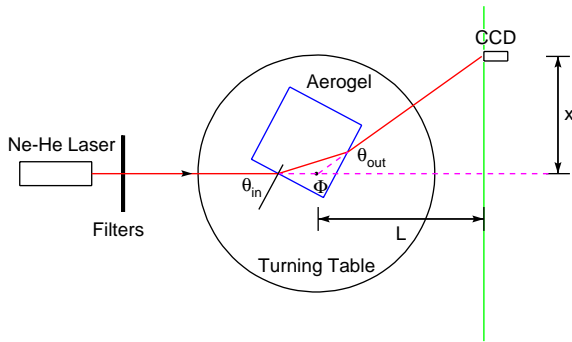


Fig. 3. Experimental set-up used for the measurement of the refractive index.

3. Behaviour with irradiation

In a RICH detector working at a pp collider [10], the aerogel can be exposed to very intense irradiation. It is therefore important to check if this can result in a deterioration of the aerogel with the consequent degradation of the particle identification performances.

Inside the LHCb detector, the aerogel wall will be positioned at a radial distance of about 10 cm [10] from the beam and at about one metre downstream of the interaction point. As a consequence, it will be exposed to a significant particle flux, up to 3.5×10^{12} particles/ cm^2 per year.

In order to investigate possible deterioration of the aerogel quality, samples of aerogel have been exposed to different sources of particles and their

optical parameters monitored at different steps of the irradiation process.

3.1. Sources of irradiation

One tile of dimensions $50 \times 50 \times 23 \text{ mm}^3$ has been exposed to a proton source (IRRAD-1 [14]) using the CERN PS T7 East Hall beam. The primary proton beam had a momentum of 24 GeV/c, the spot was $2 \times 2 \text{ cm}^2$ wide with fluxes in the range $1\text{--}3 \times 10^{13} \text{ p/cm}^2/\text{h}$. Because of the small size of the beam, irradiation was concentrated only in the center of the tile.

The exposure to IRRAD-1 was made in six steps: Table 1 summarises the fluence accumulated at each step. The total fluence amounts to 51.525×10^{12} protons/ cm^2 . The first three irradiation steps correspond each to about one year of operation of the aerogel RICH of LHCb at LHC, the last ones to larger doses that will not probably be reached in the lifetime of the detector.

Another tile of aerogel was exposed to neutrons, this time irradiating uniformly the whole tile. Neutrons were obtained using the CERN PS T8 proton beam (with momentum of 24 GeV/c) interacting on lead and carbon targets (IRRAD-2 [15]). Secondary particles in the irradiation cavity were neutrons (energy range: 50 keV–1 MeV), p , π^+ , π^- (energy range: 0.3–4 GeV) and γ (energy range: 100 keV–100 MeV). An energy cut on the spectrum (energy higher than 100 keV) was applied to calculate the dose of neutrons.

Table 1

Fluence in number of protons per cm^2 traversing the central part of the aerogel sample. For each irradiation step, the fitted values of A and C are shown. The quoted errors are those determined by the fit

Fluence (10^{12} p/ cm^2)	A (%)	C (10^{-4} $\mu\text{m}^4/\text{cm}$)
0	83.13 ± 0.04	53.8 ± 0.1
3.545	86.45 ± 0.04	54.5 ± 0.1
7.045	91.55 ± 0.04	55.2 ± 0.1
9.725	91.00 ± 0.04	55.8 ± 0.1
19.445	86.18 ± 0.04	56.9 ± 0.1
35.645	88.23 ± 0.04	54.6 ± 0.1
51.525	85.22 ± 0.04	54.5 ± 0.1

Table 2

Fluence in units of 1 MeV equivalent neutrons per cm^2 absorbed by the aerogel sample irradiated with the mixed beam (n , p , π^\pm , γ). For each irradiation step, the fitted values of A and C are shown. The quoted errors are those determined by the fit

Fluence (10^{12} n/ cm^2)	A (%)	C (10^{-4} $\mu\text{m}^4/\text{cm}$)
0	81.47 ± 0.04	57.0 ± 0.1
6.79	80.10 ± 0.04	62.3 ± 0.1
9.95	79.26 ± 0.04	64.2 ± 0.1
21.25	79.34 ± 0.04	64.7 ± 0.1
38.35	83.72 ± 0.04	67.3 ± 0.1
55.15	85.42 ± 0.04	68.3 ± 0.1

With this selection, 1 MeV equivalent neutron flux is 5.59×10^{11} n/ cm^2/h at 50 cm from the flux axis, 1.11×10^{12} n/ cm^2/h at 10 cm. The total accumulated fluence was 55.15×10^{12} n/ cm^2 , corresponding approximately to about 15 years of data taking of LHCb. Table 2 summarises the measurements at each irradiation step.

Finally, tiles of hygroscopic and hydrophobic aerogel were irradiated with γ from a radioactive ^{60}Co source used in a Gammacell² 220 unit [16] located at the “Istituto Superiore di Sanità” in Rome. The source provides an uniform irradiation by gamma rays with an energy of 1.3 and 1.7 MeV. Also in this case, the transmittance was monitored as an ageing indicator after every irradiation

²Gammacell[®] 220 is a brand name of the Atomic Energy of Canada Limited (AECL) and MDS Nordion International.

Table 3

Dose in Gy absorbed by the hygroscopic aerogel sample irradiated with γ rays from ^{60}Co . For each irradiation step, the fitted values of A and C are shown. The quoted errors are those determined by the fit. The absorbed dose is the equivalent dose calculated for SiO_2

Dose (Gy)	A (%)	C (10^{-4} $\mu\text{m}^4/\text{cm}$)
0	97.09 ± 0.04	44.4 ± 0.1
38296	97.38 ± 0.04	44.0 ± 0.1
73364	93.37 ± 0.04	39.0 ± 0.1
115654	98.24 ± 0.04	41.5 ± 0.1
229081	98.28 ± 0.04	41.0 ± 0.1

Table 4

Dose in Gy absorbed by the hydrophobic aerogel sample irradiated with γ rays. For each irradiation step, the fitted values of A and C are shown. The quoted errors are those determined by the fit. The absorbed dose is the equivalent dose calculated for SiO_2

Dose (Gy)	A (%)	C (10^{-4} $\mu\text{m}^4/\text{cm}$)
0	94.33 ± 0.3	78.9 ± 0.2
37305	96.59 ± 0.3	77.0 ± 0.2
72373	98.18 ± 0.5	61.3 ± 0.2
114663	97.99 ± 0.5	79.4 ± 0.2
228090	98.36 ± 0.3	71.0 ± 0.2

period. Five irradiation steps provided a total dose of ≈ 230 kGy, corresponding to the dose absorbed by the innermost region of the aerogel radiator in ~ 30 years of operation of LHCb. Table 3 summarises the measurements with the hygroscopic aerogel, and Table 4 those concerning the hydrophobic aerogel. The results with γ irradiation on hydrophobic aerogel agrees with previous tests [17].

3.2. Measurements and results

The transmittance of the irradiated tiles was measured before and after each irradiation step, and the curves were then fitted with the Hunt equation (1) to determine the A and C parameters. During irradiation, the hygroscopic tiles were sealed in a polyethylene bag filled with nitrogen

to ensure isolation from humid air. Each time the transmittance was measured, the aerogel block was exposed to air for about one hour.

In order to separate effects due to the handling of the tiles, measurements were made systematically on a reference tile, with optical parameters A and C very close to those of the irradiated ones. The comparison of the two samples was then made to account for the effects due to handling in the irradiation measurements. The ratio of the transmittances of the irradiated and the reference samples is plotted in Fig. 4 for different wavelengths as a function of the absorbed dose. Fig. 5 presents the ratios for the clarity. For proton irradiation, no evidence of degradation of the aerogel optical parameters is visible: both parameters A and C are independent of the fluence.

The results from the neutron exposure show a more complex response. While T does not change for λ above 400 nm, the value of the clarity factor C has an increasing trend, as if the structure of the silica aggregates and empty bubbles inside the tile were modified by the high flux of particles traversing the aerogel sample. The worsening in the clarity factor is mainly determined by the behaviour at small values of wavelength as can be seen from the transmittance ratios in Fig. 4b.

No detectable decrease of the aerogel optical quality has been found under gamma irradiation, as shown in Fig. 6: the transmittance T is unchanged within the experimental uncertainties. For this kind of irradiation, the index of refraction was measured in the initial conditions and after $\simeq 20000$ Gy. Also for this parameter no

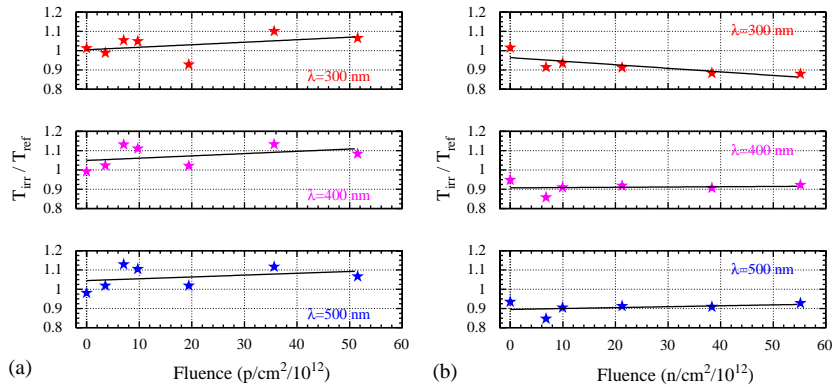


Fig. 4. Transmittance ratios $T_{\text{irr}}/T_{\text{ref}}$ as a function of the absorbed dose at different wavelengths with (a) protons and (b) neutrons. A linear fit has been superimposed.

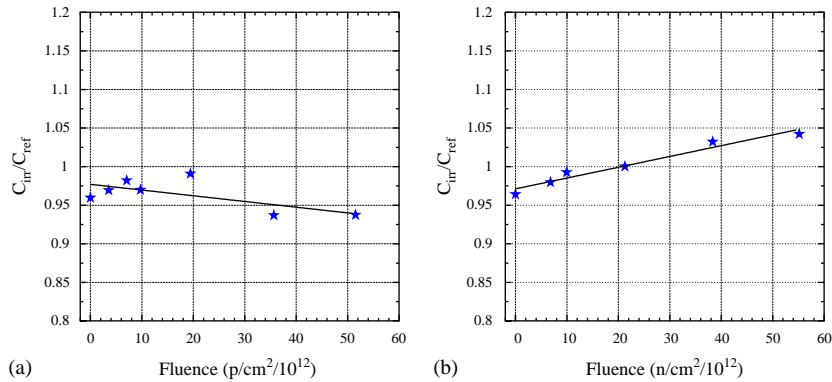


Fig. 5. Clarity ratios $C_{\text{irr}}/C_{\text{ref}}$ as a function of the absorbed dose with (a) protons and (b) neutrons. A linear fit has been superimposed.

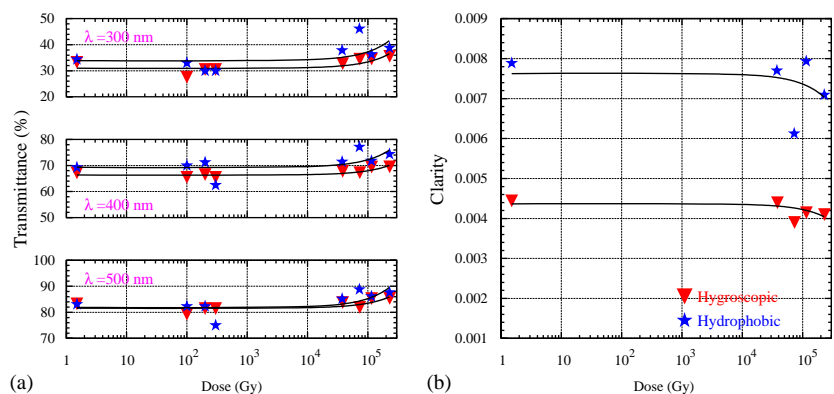


Fig. 6. (a) transmittance as a function of the absorbed dose of γ at different wavelengths; (b) clarity as a function of the absorbed dose. A linear fit has been superimposed.

Table 5
Index of refraction n for hygroscopic and hydrophobic aerogel samples as a function of the absorbed dose of γ at $\lambda = 632.8$ nm
Errors are described in Section 4.2.

Absorbed dose (Gy)	Hygroscopic	Hydrophobic
0	1.0336 ± 0.0003	1.0297 ± 0.0003
18797.0	1.0326 ± 0.0003	—
19788.0	—	1.0304 ± 0.0003

detectable variations have been observed, as shown in Table 5. No comparison with reference tiles was available in the case of γ irradiation.

4. Behaviour with humidity

Hygroscopic aerogel can absorb water vapour, forming OH groups. These can be eliminated by baking the aerogel at high temperatures. The humidity test was performed using the following procedure. The aerogel tile was baked at a temperature of $\simeq 500^\circ\text{C}$ for about 4 h. That temperature was reached in about five hours in order to avoid possible cracks due to thermal stress. At the end of this first baking process, the index of refraction and transmittance were measured and A and C were determined.

The aerogel block was then exposed to humid air for about one week; the optical parameters were remeasured. Finally, a new baking cycle was

done in order to find out if it was possible to restore the initial values.

4.1. The experimental set-up

The setup used to expose aerogel to humid air was similar to a greenhouse. Fig. 7 shows the set-up: the aerogel tile was placed on a high precision balance. Glasses of water were placed near the tile, inside the transparent box, creating a humid environment. The temperature of the room was about 24°C , and the variations of the aerogel weight caused by the absorption of water were recorded by the balance as a function of temperature and of relative humidity which were monitored with a computer and are shown in Fig. 8. This figure shows also the variation of the aerogel weight as a function of time. The steep rise indicates that at the beginning the aerogel can absorb a large amount of water vapour, then the absorption of water slows down. Oscillations are due to a day/night effect in the temperature of the room.

The index of refraction n and light transmission T of the hygroscopic aerogel were measured as a function of the humidity absorbed by the tile. If the relative humidity of the room (where measurements were done) is different from the one of the aerogel environment inside the box, the aerogel can absorb or expel some water to reach equilibrium. This is a quite fast process, so in order to



Fig. 7. The experimental set-up used for the humidity test. The aerogel is located on the plate of the balance, the atmosphere inside is saturated with vapour of water.

keep the aerogel under uniform conditions and minimize variations of weight, measurements were made turning off the air conditioning system of the room, inducing a relative humidity between 60% and 70%.

4.2. Measurements of the refractive index

The prism method, as described in Section 2.2, provides an accurate measurement of n . The uncertainty on n is related to that of the two angles Φ and θ_{out} ; this latter is determined by the two distances x and L , as shown in Fig. 3. From formula (2), the error on the refractive index is given by

$$\sigma_n^2 = \left(\frac{\partial n}{\partial \Phi}\right)^2 \sigma_\Phi^2 + \left(\frac{\partial n}{\partial x}\right)^2 \left[\sigma_x^2 + \frac{x^2}{L^2} \sigma_L^2\right]. \quad (3)$$

The laser beam spot on the CCD display presents a central region about 2 mm wide with

Table 6

Index of refraction n measured with the green laser source as a function of weight. Spreads in the measurements are also shown

m	8.06 g	8.09 g	8.35 g	8.63 g
n	1.0335	1.0331	1.0345	1.0363
δn	0.0005	0.0009	0.0012	0.0029

halos around it, as shown in Fig. 9. The center of gravity of the image was used to measure the x displacement on the screen. The precisions on x and L are $\sigma_x \cong 0.5$ mm and $\sigma_L \cong 5$ mm, respectively. The uncertainty on the angle Φ is essentially determined by the small deviation from $\Phi = \pi/2$ due to local imperfections on the surfaces of the tiles crossed by the laser beam. Assuming $\sigma_\Phi = 0$, the uncertainty on n is $\sigma_n \cong 0.0003$. The contribution to σ_n due to the uncertainty on the angle Φ is plotted in Fig. 10. The contribution to σ_n due to the definition of the minimum displacement condition is negligible ($\sigma_n < 7 \times 10^{-5}$).

The refractive index was then determined from several measurements corresponding to different entrance points on the aerogel. The mean value was taken as the final measurement.

Table 6 lists the values of n as a function of the absorbed humidity reflected in the increase of the aerogel weight. The spreads δn (R.M.S.) of each set of measurements are also shown. These spreads are related to local non-homogeneities of the tiles, but they are also connected to the fast variation of the aerogel weight during the measurement procedure (as explained in Section 4.1). The refraction index n increases with the amount of absorbed water vapour. The maximum variation of $\Delta n/n$ (corresponding to an increase of mass of 6.7%) was 0.3% for the green laser source. Such a kind of variation would give a large contribution to the performances of particle identification with RICH detectors.

4.3. Measurement of transmittance

Fig. 11 shows the results for the transmittance T at different wavelengths and for the clarity factor C measured at different steps of water absorption. A linear fit has been superimposed. For C an

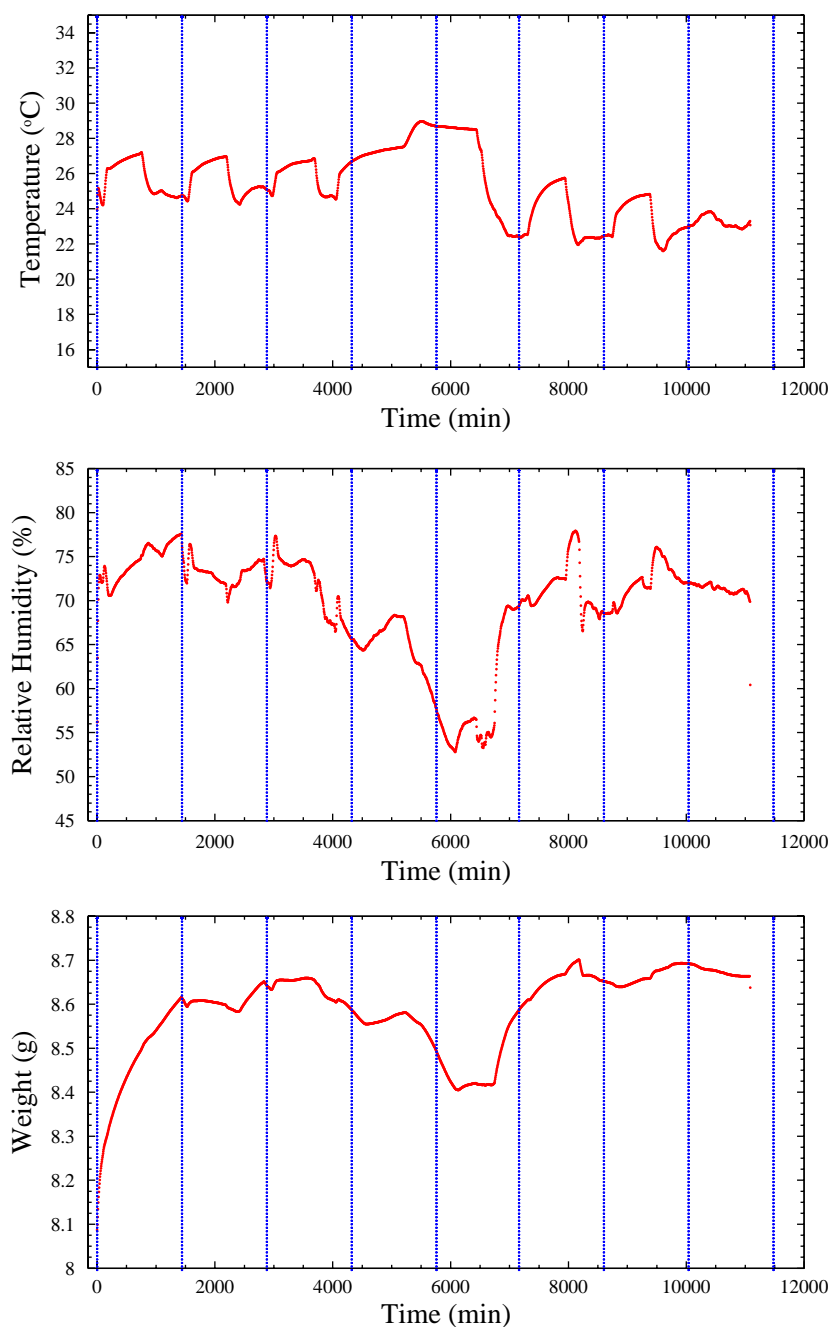


Fig. 8. Temperature (°C) and relative humidity (%) of the room and weight (g) variation of the aerogel tile as a function of time (in minutes); vertical lines indicate intervals of 24 h.

increase as a function of the absorbed humidity is measured in the data. Table 7 summarizes the results of the measurements.

It must be noted that the aerogel tiles of the LHCb RICH detector will be encapsulated in an hermetic box, and therefore the exposure to

humidity will be certainly lower than the one considered for this test.

5. Conclusions

Ageing effects on aerogel due to irradiation with different types of incident particles and due to humidity absorption were investigated by monitoring the optical parameters characterising the quality of the aerogel tiles. The parameter A , the clarity factor C and the index of refraction n were measured.

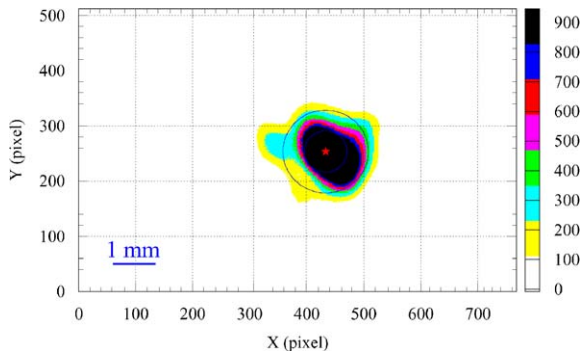


Fig. 9. Typical picture of the laser beam spot from the CCD. The star indicates the center of gravity of the image; 1 and 2 mm diameter circles are also plotted.

The performed measurements show no evidence of ageing of the aerogel when irradiated with protons and γ , and a moderate degradation of the clarity when irradiated with neutrons. On the other hand, the humidity test revealed that a prolonged exposure to humid air does not irreversibly degrade the properties of the aerogel, which are completely restored by baking the exposed sample at $\approx 500^\circ\text{C}$.

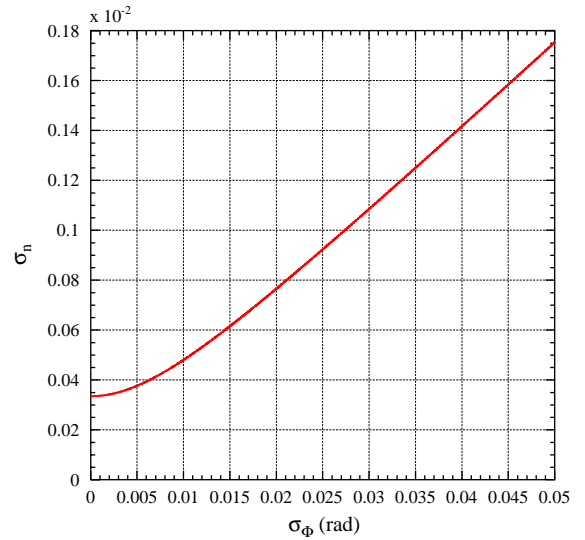


Fig. 10. The error σ_n as a function of the uncertainty on the angle Φ at the tile vertex when $\sigma_x = 0.5$ mm and $\sigma_L = 5$ mm.

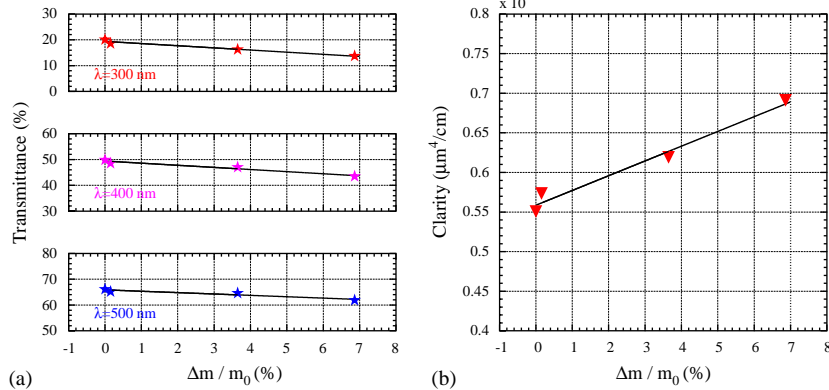


Fig. 11. (a) transmittance as a function of $\Delta m/m_0$ (%) at different wavelengths during the humidity test; (b) clarity as a function of $\Delta m/m_0$ (%).

Table 7

Values of the parameters A and C as a function of weight for different steps in the humidity test

m	8.06 g	8.07 g	8.35 g	8.61 g
A (%)	82.49 ± 0.04	81.94 ± 0.04	82.54 ± 0.04	81.35 ± 0.04
C (10^{-4} $\mu\text{m}^4/\text{cm}$)	55.1 ± 0.1	57.3 ± 0.1	61.9 ± 0.1	69.1 ± 0.1

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