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Preliminary study of silica aerogel as a gas-equivalent material in ionization chambers

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ABSTRACT

Since about two decades, a renewed interest on aerogels has risen. These peculiar materials show fairly unique properties. Thus, they are under investigation for both scientific and commercial purposes and new optimized production processes are studied. In this work, the possibility of using aerogel in the field of radiation detection is explored. The idea is to substitute the gas filling in a ionization chamber with the aerogel. The material possesses a density about 100 times greater than ambient pressure air. Where as the open-pore structure should allow the charge carriers to move freely. Small hydrophobic silica aerogel samples were studied. A custom ionization chamber, capable of working both with aerogel or in the classic gas set up, was built. The response of the chamber in current mode was investigated using an X-ray tube. The results obtained showed, under proper conditions, an enhancement of about 60 times of the current signal in the aerogel configuration with respect to the classic gas one. Moreover, some unusual behaviours were observed, i.e. time inertia of the signal and super-/sub-linear current response with respect to the dose rate. While testing high electric fields, aerogel configuration seemed to enhance the Townsend's effects. In order to represent the observed trends, a trapping–detrapping model is proposed, which is capable to predict semi-empirically the steady state currents measured. The time evolution of the signal is semi-quantitatively represented by the same model. The coefficients estimated by the fits are in agreement with similar trapping problems in the literature. In particular, a direct comparison between the benchmark of the FET silica gates and aerogel case endorses the idea that the same type of phenomenon occurs in the studied case.

1. Introduction

In the last two decades a renewed interest in aerogels has risen [1–4]. The unique properties of this class of materials (*i.e.*, extremely low density, high dielectric constant, high specific surface area *etc.*) make them interesting for both commercial and scientific applications. In this frame, the optimization of production processes and the control of chemical–physical characteristics of the aerogels are becoming of great interest. In this paper, we try to explore a new application of aerogels in radiation measurements.

The idea is to exploit both the *open-pores* characteristic of aerogel and the higher density of the material (around 0.1 g cm^{-3} in the application described here), if compared to ambient pressure gas, in order to obtain a *gas equivalent* filling material for an ionization chamber (IC). The higher density permits to increase the primary interactions and energy deposition (as in pressurized IC) and the open pores structure allows the transport of charge carriers. The response in current mode of a custom IC was then tested in both gas configuration and aerogel configuration (AIC), in order to verify the effective capability of the aerogel to be used as a gas equivalent material.

Different samples of hydrophobic silica aerogel were tested in both air and argon atmospheres, and the response was explored as a function of both the bias voltage and the dose rate. The irradiations have been performed with an X-ray tube at the secondary standard calibration laboratory at the Department of Energy at the Polytechnic of Milan.

2. Experimental set up

The custom IC consists of an aluminium box $11.5 \times 9 \times 5$ cm^3, with wall thickness of about 2 mm. Two thin brass screw supports are used to maintain alignment of the two brass plates. The plates are circular 2 mm thick, with different radii. With reference to Fig. 1, the upper

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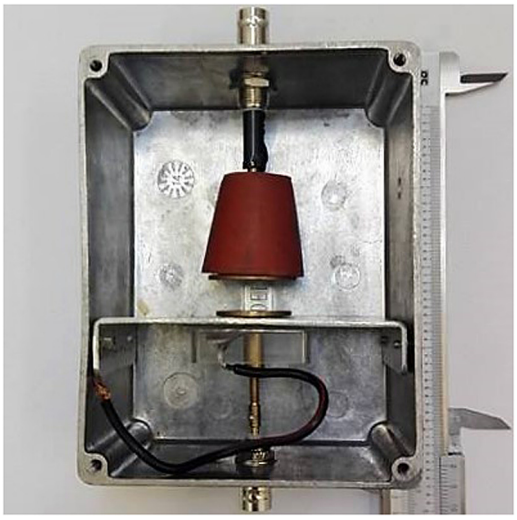

Fig. 1. Frontal view of the IC. During measurements, the IC is closed by putting the aluminium cover on the front face of the chamber.

Table 1
Specifications of N.A.M. Srl. aerogel samples.

Specific	Value	Unit
Colour	Opaque white	–
Density	0.09 ± 5%	g/ml
Specific surface area	889 ± 30	m^2/g
Mean pore diameter	~15	nm
Total porosity volume	~96	%
Behaviour with H_2O	Hydrophobic	–

one is 1.35 cm and the bottom one is 1.25 cm. This configuration permits to enlarge or tighten the sensitive volume between the plates (the distance between plates can be tuned in the range 0.4–1.6 cm) by screwing/unscrewing the electrodes. It also makes possible to insert or to remove the aerogel samples. The larger plate is kept at high voltage, while the other one is connected to a picoammeter Keithley model 6517. Surrounding the smaller plate, a 2 mm thick aluminium guard ring, with a circular hole of the same size of the larger plate, is equipotential to the box. A rubber pierced plug surrounds the high voltage support in order to avoid discharges between the support and the box while high voltage is applied. A small plastic support, covered with a thin copper foil equipotential to the box, is attached on the lower face of the low potential electrode. The copper foil permits to kill the electric field below the smaller electrode as shown in Fig. 2. The box has a gas inlet for working in continuous argon flow.

The aerogels samples were provided by N.A.M. Srl.[1] The dimensions of the aerogel samples are $2.5 \times 2.5 \times 0.4$ cm^3 and $2.5 \times 2.5 \times 0.8$ cm^3. Specifications and composition of the material are reported in Tables 1 and 2.[2]

The silica precursors used in the production phase were tetramethoxysilanes (TMOS). As the aerogels prepared from TMOS show hydrophilic characteristics [6], trimethylchlorosilanes (TMCS) were added to the mother solution during ageing. The hydrophilic –OH groups are substituted by hydrophobic organic radicals, and so the final product becomes hydrophobic. Besides, the absence of electronegative species like –OH permits the electron transport through the aerogel.

[1] N.A.M. Srl (Nano Analysis and Materials) is an Italian start-up and an accredited spin-off of the University of Pavia [5].

[2] The determination of the composition was obtained by ICP Mass Spectroscopy made at the Polytechnic of Milan—Dep. of Energy radiochemistry laboratory, with the following procedure. Small pieces of aerogel were dissolved in acid mixture inside a microwave oven. From the liquid solution aerosols were extracted and introduced in the plasma for spectroscopy.

Table 2
N.A.M. Srl. silica aerogel chemical composition in weight percentage (w/w) and oxides. LOQ is the Limit Of Quantification.

Species	$\%_{w/w}$	$\%_{oxide}$
SiO_2	85	–
$–CH_3$ groups	15	–
Na_2O	0.0082 ± 0.0003	0.0110
MgO	0.0007 ± 0.0001	0.00116
Al_2O_3	0.1363 ± 0.0008	0.2574
K_2O	<0.139 (LOQ)	<0.1675
CaO	<0.140 (LOQ)	<0.1960
Fe_2O_3	<0.00113 (LOQ)	<0.00186

After a proper gelation and ageing time, the wet gels were put in an autoclave (super-critical drying method [1,4]) and finally the dried gel was obtained.

The brittleness of the material required a careful handling. In particular, the smaller samples were found to be excessive fragile while they were put inside the chamber, and almost all the measurements were made with the thicker samples.

3. Measurements

The measurements were made using the ion chamber in four configurations, in particular:

1. Gas configuration, air atmosphere (IC-AIR)
2. Aerogel configuration, air atmosphere (AIC-AIR)
3. Gas configuration, argon atmosphere (IC-AR)
4. Aerogel configuration, argon atmosphere (AIC-AR)

In the case of gas configuration, the distance between plates was set to 8 mm, equivalent to the thickness of the aerogel samples. The bias voltage was tuned from 0 up to 5000 V.

Irradiation was performed using two different X-ray beams: i) the standard ISO-4037 W300 radiation quality (mean energy 199 keV, maximum dose rate 1.4 Gy h^{-1}at 1 m from the source) and ii) a non standard beam produced using the added filtration of the W200 quality with a H.V. at the X-ray tube of 300 kV (in the following called NSW300). The NSW300 was simulated with the MCNPX (version 2.6.0) [7] Monte Carlo code, finding a mean photon energy of about 194 keV. The experimental kerma rate at maximum X-ray tube current was measured to be 4.72 Gy h^{-1}at 1 m.

The beam illuminated the chamber perpendicular to the front face, with reference to Fig. 1, at the fixed distance of 1 m from the source. The dose rate was changed by varying the X-ray tube current from 0 up to 10 mA.

3.1. Measurements in air atmosphere

The instrument was firstly characterized in the classic gas configuration in air (IC-AIR). In particular, the attention was focused on the parasitic electric field effects, which are supposed to be present. Actually, the overall layout of the instrument is not optimized with respect to parasitic effects, as the main issue is the possibility to insert and remove safely the aerogel samples.

In the IC-AIR configuration a mean plateau current of about 75–80 pA was measured at 10 mA tube current, slightly increasing while the bias voltage was raised. The measured signal was then compared to the expected theoretical current value, in order to evaluate both the parasitic electric field effects and the volume recombination contributions.

The theoretical saturation current was calculated using the formula [8]

$$I_{sat} = N_0 d S e \quad (1)$$

where N_0 is the *ionization density* (ions cm^{-3} s^{-1}), which was determined using the MCNPX Monte Carlo code. d and S are the distance between