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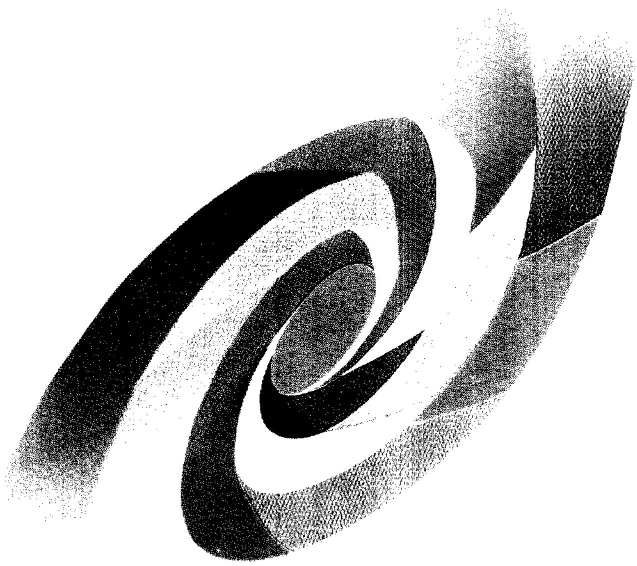

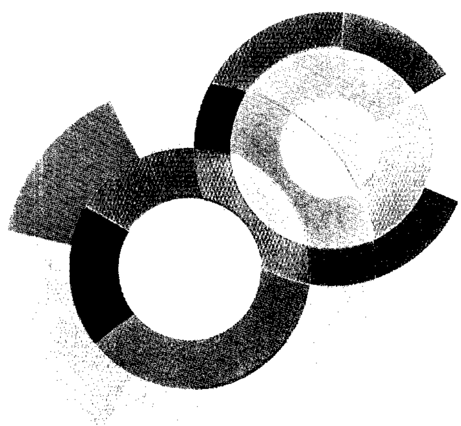

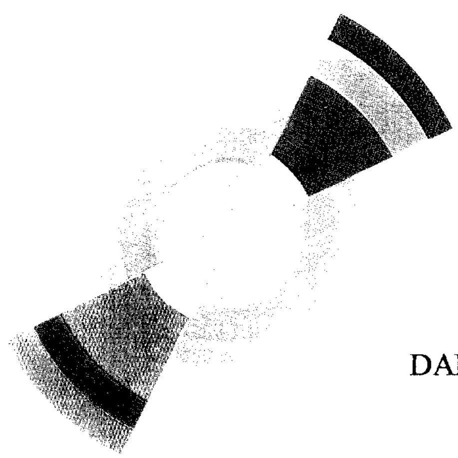

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SOL-GEL COATING OF SCINTILLATING CRYSTALS

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Sol-gel coating of scintillating crystals

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Abstract

We propose a new method for the coating of scintillating crystals that combines low thickness and optical efficiency. In this method, the air-gap between crystal and wrapping is replaced by a sol-gel optical coating of very low refractive index (< 1.30). The external wrapping is replaced by a diffusive or reflective layer, which can also be made by the sol-gel technique. This leads to a thin coating of a few microns, with light collection properties comparable to usual wrapping, good mechanical and radiation resistance.

Résumé

Nous proposons une nouvelle méthode pour le revêtement des cristaux scintillants, combinant faible épaisseur et efficacité optique. Dans cette méthode, l'épaisseur d'air entre cristal et habillage est remplacée par un revêtement optique sol-gel de très faible indice de réfraction (< 1.30). L'habillage externe est remplacé par une couche réflectrice ou diffusante, qui peut être également réalisée par la technique sol-gel. On obtient ainsi un revêtement mince de quelques microns, possédant des propriétés de collection de lumière comparables aux habillages usuels, des bonnes résistances mécanique et aux radiations.

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1 Introduction

In scintillating crystals, only a small amount of the emitted light reaches directly the photodetector. Most of the light encounters one or more reflections on lateral surfaces before being detected, specially in long crystals. In the case of a long bare crystal, the light should be in total internal reflection to have a reasonable chance to reach the photodetector.

Usually the light collection is improved by coating the surfaces. Two types of solutions are used. In the first, either a diffusive film, like white paint, or a reflective metallic layer, is deposited onto the surface. The crystals are optically isolated by a low thickness film. However the total internal reflection is not conserved, and the yield could be worse, particularly in long crystals where many reflections occur. In the second type, the crystals are wrapped in white diffusive sheets of paper, Tyvek®, Teflon®, or any non-absorbing material. In that case the total reflection is conserved due to the air gap between crystal and wrapping. For example improvement of about 30 % in light yield, compared to a bare crystal, can be obtained in lead tungstate covered by Tyvek. This improvement is paid by a loss in compactness.

In an intermediate solution, the air-gap is replaced by an optical coating, with the advantage to combine low thickness with at least a part of total reflection. However, to keep enough light in total reflection, this coating should have an index of refraction sufficiently low, less than 1.3, and a thickness of more than one micron. For these reasons, the classical deposition techniques, such as vacuum evaporation, CVD, etc., are not satisfactory. They produce indices of refraction barely less than 1.4, and thicknesses over a few tenths of micron are difficult to obtain. We have solve these problems using a sol-gel deposition technique which allows the deposition of optical film with very low index and good mechanical resistance [1].

We will in a first part describe the sol-gel process developed, then show light yield characteristics of some lead tungstate crystals treated with that process and compare them with the usual Tyvek wrapping.

2 The sol-gel process

They are two main procedures for the production of optical coating by sol-gel. The colloidal route starts from a precursor solution, which is transformed in a colloidal suspension containing metallic oxide particles, and gives after deposition porous and soft layers, with very low refractive indices (down to 1.22) and poor mechanical resistance. The polymeric route uses a polymer solution, which after deposition and densification gives a dense and hard film. We have used a combination of this two processes, developed at CEA/Limeil and called the "sol-silox" preparation, which is a colloidal silica sol grafted by a siloxane [3].

As shown in Table 1, the proportion of colloid versus binder acts directly on the index value and the mechanical strength. We have choose in this application a ratio 70 – 30, which gives a good compromise between index and abrasion resistance.

Table 1: Variation of the index of refraction in function of the colloid / binder ratio in the sol-silox process [3].

colloid/binder ratio	refractive index
100 – 0	1.22
90 – 10	1.22
80 – 20	1.22
75 – 25	1.23
70 – 30	1.26
60 – 40	1.38
50 – 50	1.40

We have treated lead tungstate crystals whom characteristics are : tapered shape, dimensions $\approx 17.5 \times 225 \times 20.3$ mm^3, weight ≈ 1 kg. The film is deposited on the four lateral faces using a standard dip coating process followed by a low temperature curing at 200°C during 1/2 h. This cycle gives a 0.2 micron thick, inert film, and should be repeated 4 to 6 times to obtain a total thickness of 1 micron. The abrasion resistance of the coating is in compliance with the test 'US-MIL-C-0675 moderate'.

We have also tested the resistance to radiation of these coatings. 700 nm thick films, identical in composition to those deposited on crystals, have been deposited by spin coating on silica plates. They have been submitted to a gamma dose up to 30 kGy (equivalent in air) in our ^{60}Co irradiation facility Cocase, at a dose rate of about 200 Gy/h, and compared to uncoated plates exposed to the same dose. As shown in figure 1, after correction of the substrate absorption, only little degradation, less that 3 %, is seen below 250 nm, out of the range of emission of almost all scintillators.

After the sol-gel deposition, a silver reflective film was deposited on the side faces of the crystal, by a standard vacuum evaporation technique. To prevent oxidisation of the silver film a protective polymeric layer was deposited on some of the crystals.

3 Light yield results

A first, early, crystal (#684) has been measured using the cosmic ray test bench developed in Saclay for CMS crystal monitoring R&D [2]. This test bench gives access to the absolute photoelectron response of the crystal

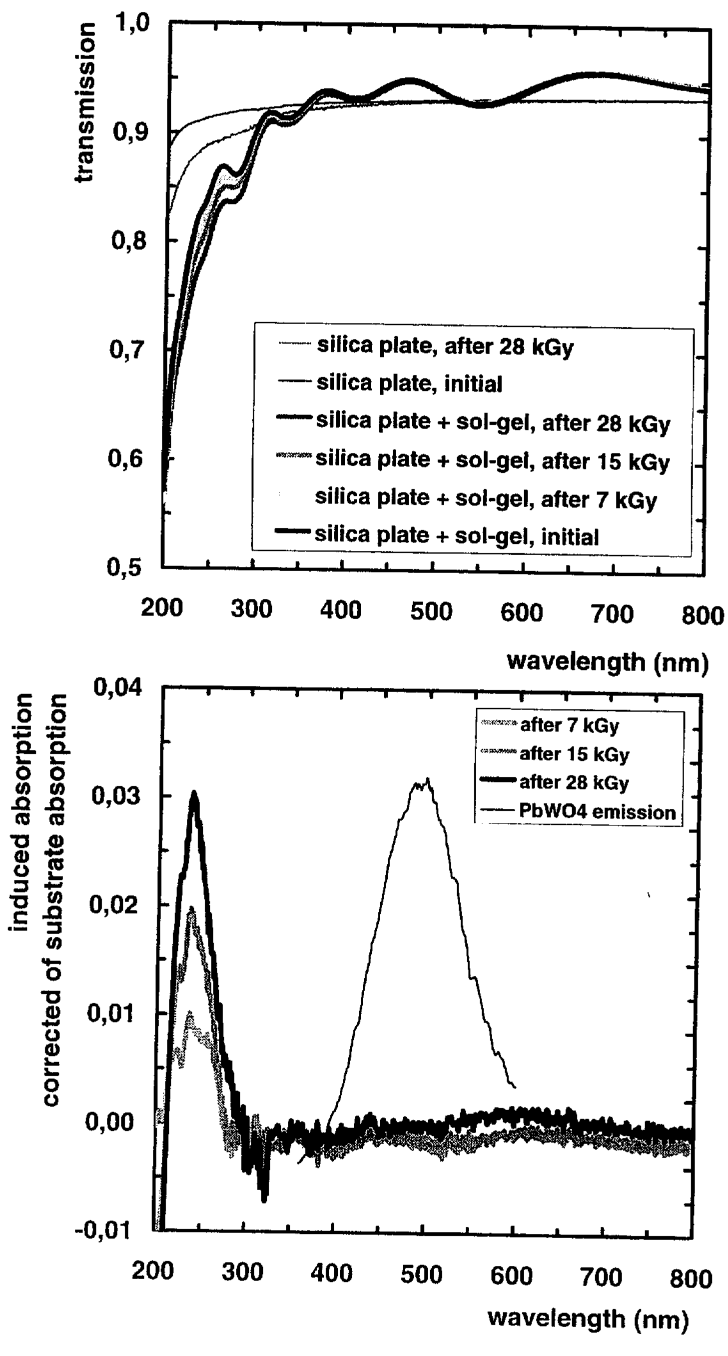

Figure 1: Variation under gamma radiation of the optical transmission of silica plates covered by 700 nm thick sol-silox films, compared to uncoated silica plates (upper) ; induced absorption of the sol-gel film, after correction of the silica substrate absorption (lower). The emission spectra of lead tungstate is recalled in arbitrary units, for wavelength range comparison.

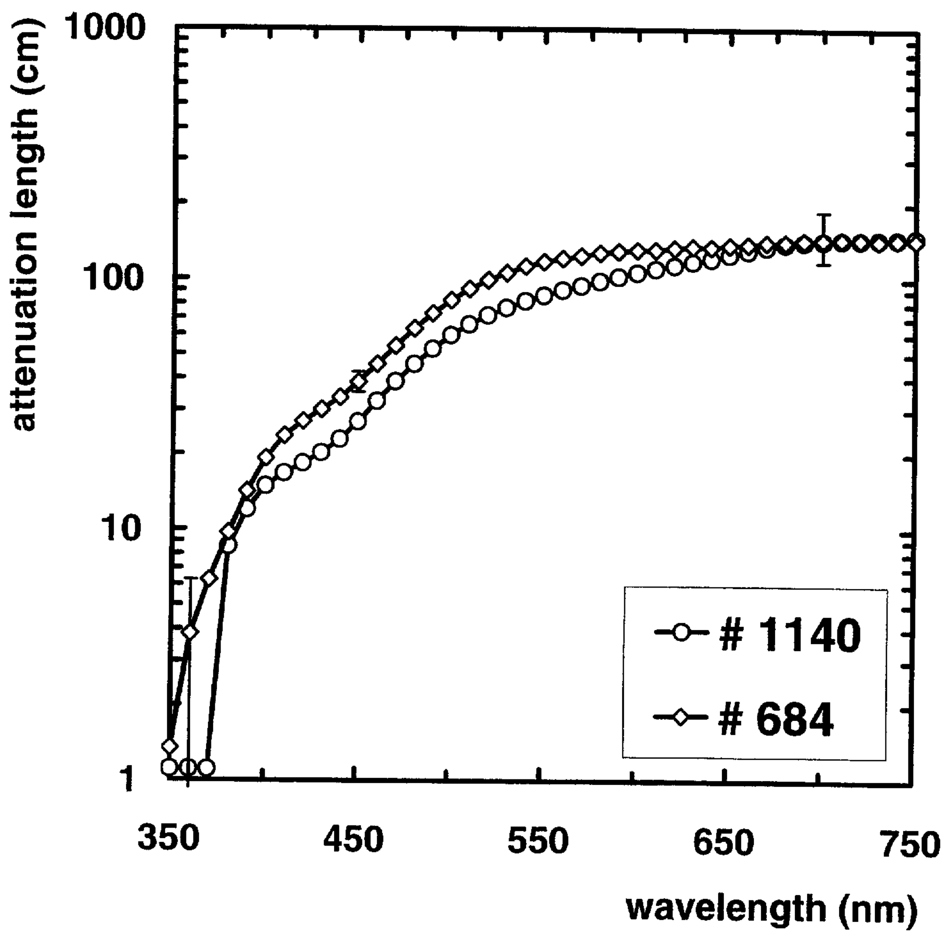

Figure 2: Attenuation length of $PbWO_4$ crystals #684 and #1140, measured longitudinally.

in function of the z position in the crystal. The signal is corrected for temperature variation and crystal size effect.

The optical transmission of the crystal is shown in figure 2. The crystal has been measured before sol-gel treatment, with the following types of coating : nothing ("bare crystal" on figure), wrapped with Tyvek on lateral faces, with ("Tyvek+mirror") or without ("Tyvek on lateral faces") an aluminized Mylar reflective sheet at the end of the crystal opposite to the photodetector, (called front end hereafter) and after sol-gel treatment, with ("sol-gel+Ag+mirror") the same reflector at the front end.

Figure 3 shows the signals for the different coatings (normalized to the signal at $z = 8$ cm for the bare crystal). In this crystal, the light yield with sol-gel coating is comparable to the one with Tyvek, 45 % over the bare crystal response and slightly below (15 %) the full (Tyvek+mirror) coating. The influence of reflectors at crystal extremities, which is known to be an important factor in the light yield and light yield evolution [4], should be noted.

A second crystal (#1140) has been measured using the automatic crystal control system (ACCOS prototype 0) developed in Annecy in which the light yield in function of the position is measured with the help of a ^{22}Na source by the so-called "start/stop" method [5, 6].

Similarly to the preceding one, the crystal has been measured before sol-gel treatment, with the following types of coating : nothing ("bare crystal" on figure), Tyvek over faces, including or not the front end (respectively "Tyvek" and "Tyvek on lateral faces"), and after treatment, with ("sol-gel+Ag+mirror"), or without ("sol-gel+Ag") reflector at the front end. Figure 4 shows the signals for the different coatings. In this crystal, the light yield with sol-gel coating is comparable to the one with the full Tyvek coating, 40 % over the bare crystal response. Again, the influence of reflectors at extremities is shown, independently of the treatment of lateral faces.

These results show that the important gain in coating thickness induced by the sol-gel+reflector coating is not counterbalanced by a light yield decrease. The light collection efficiency is similar to the one achieved with standard diffusive wrappings.

The weak point of the process is the need of a reflective layer (here silver), which presents two disadvantages : it uses a vacuum deposition technique, not needed for sol-gel itself, and it should be protected against oxidisation. We have proposed to replace this layer by a sol-gel diffusive film, based on a similar process using high index materials such as TiO_2. This would lead to a complete coating with a single, soft, technology.

However, the choice by the CMS collaboration of an alveolar structure for the lead tungstate crystals of it's electronic calorimeter [7], without any wrapping of crystals, the role of optical reflection being assumed by the structure itself, reduces the interest of the sol-gel solution, and we have not continued our developments in this field.

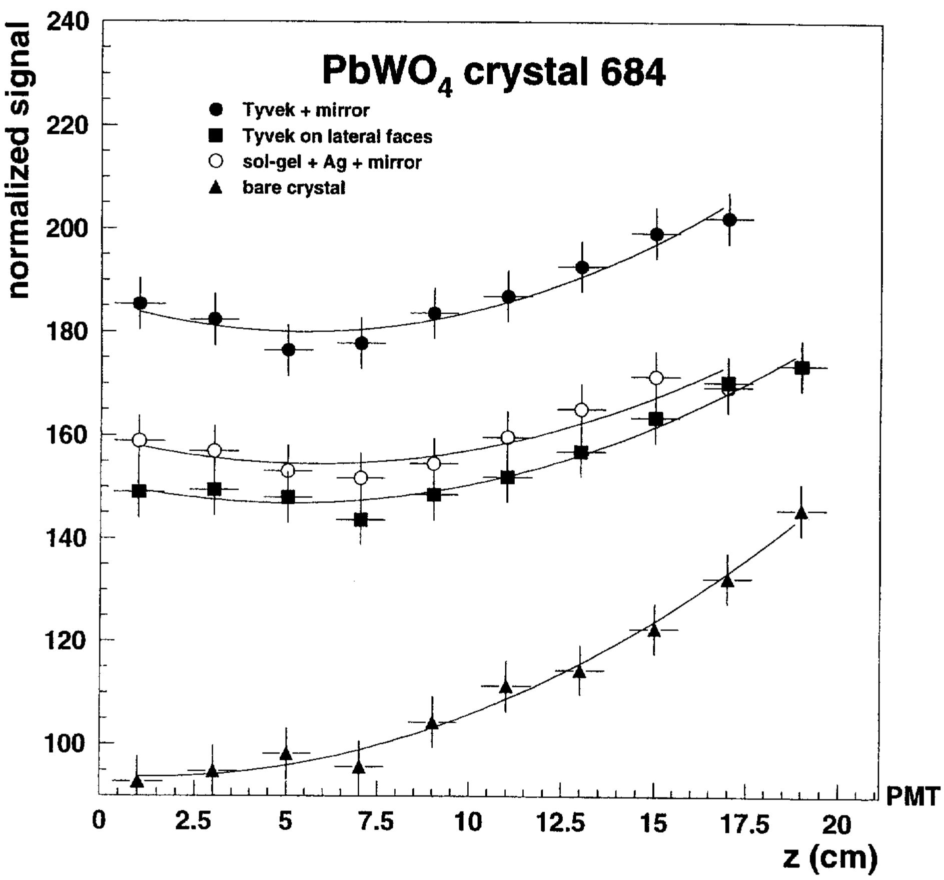

Figure 3: Response to cosmic rays of $PbWO_4$ crystal #684 with various coating types. Data are normalized to the response for the bare crystal at $z = 8$ cm. (z is the distance from the face opposite to the PMT).

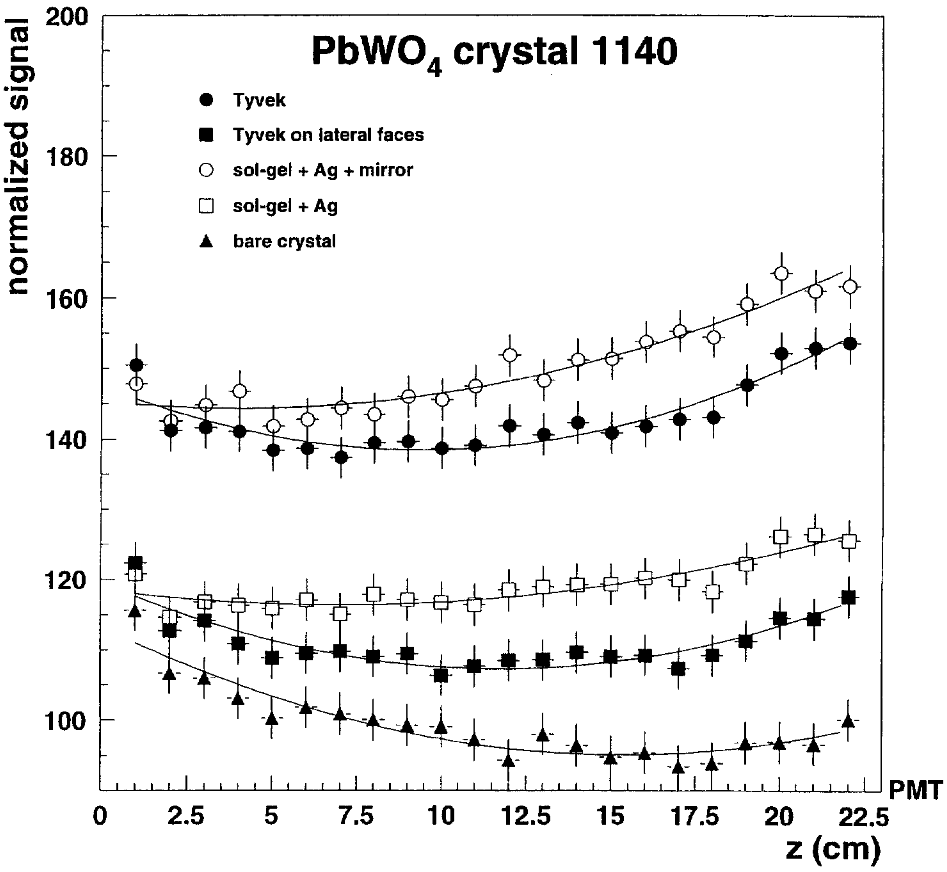

Figure 4: Response to ^{22}Na source for $PbWO_4$ crystals #1140 with various coating types. Data are normalized to the response for the bare crystal at $z = 8$ cm. (z is the distance from the face opposite to the PMT).

4 Conclusion

The sol-gel process allows the deposition on scintillating crystals of optical layers with a very low index of refraction (< 1.30). One of its advantages is that it is a "soft" process, which do not require high vacuum or high temperature techniques. The external wrapping is replaced by a diffusive or reflective layer, which might eventually also be made by the sol-gel technique. This leads to a thin coating of a few microns, with light collection properties comparable to standard wrapping, good mechanical and radiation resistance. This process allows the production of matrices of scintillators with very thin inactive layers and good light collection. It is presently considered for medical imaging applications [8].

5 Acknowledgements

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