

Progress in coupling MPGD-based Photon Detectors with Nanodiamond Photocathodes

F. M. Brunbauer^f, C. Chatterjee^a, G. Cicala^d, A. Cicuttin^e, M. L. Crespo^e, D. D'Ago^b, S. Dalla Torre^a, S. Dasgupta^a, M. Gregori^a, S. Levorato^a, T. Ligonzo^c, M. Lisowska^{f,g}, M. S. Leone^d, R. Rai^e, L. Ropelewski^f, F. Tessarotto^{a,*}, Triloki^a, A. Valentini^c, L. Velardi^d

^aINFN Trieste Trieste Italy

^bUniversity of Trieste and INFN Trieste, Trieste, Italy

^cUniversity Aldo Moro of Bari and INFN Bari, Bari, Italy

^dCNR-ISTP and INFN Bari, Bari, Italy

^eAbdus Salam ICTP, Trieste, Italy and INFN Trieste, Trieste, Italy

^fEuropean Organization for Nuclear Research (CERN), CH-1211 Geneva 23, Switzerland

^gWrocław University of Science and Technology, Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland

Abstract

The next generation of gaseous photon detectors is requested to overcome the limitations of the available technology, in terms of resolution and robustness. The quest for a novel photocathode, sensitive in the far vacuum ultra violet wavelength range and more robust than present ones, motivated an R&D programme to explore nanodiamond based photoconverters, which represent the most promising alternative to cesium iodine. A procedure for producing the novel photocathodes has been defined and applied on THGEMs samples. Systematic measurements of the photo emission in different Ar/CH₄ and Ar/CO₂ gas mixtures with various types of nanodiamond powders have been performed. A comparative study of the response of THGEMs before and after coating demonstrated their full compatibility with the novel photocathodes.

Keywords: Hydrogenated nanodiamond photocathode, Gaseous Photon detectors, MPGD, THGEM

1. Introduction

Gaseous Photon Detectors (PDs) have played an essential role in RICH applications and represent one of the best options for covering large areas with photosensitive detectors. They are not commercially available but they can be produced with a moderate investment; their operation is compatible with the presence of a magnetic field and they can offer a low material budget. The first gaseous PDs operated with photo-ionizing vapours, but CsI photocathodes are currently used in almost all applications.

CsI-based gaseous PDs have a high Photon Detection Efficiency (PDE) in the wavelength range below 200 nm. CsI is more robust than other photoconverting materials used in vacuum-based PDs but, in spite of its widespread use and successful applications, it presents limitations due to its hygroscopic nature. A degradation in Quantum Efficiency (QE) appears in case of water vapour absorption [1] making the handling of CsI photocathodes a very delicate operation. QE degradation also appears after intense ion bombardment, when the integrated charge is ~ 1 mC/cm² [2] or larger. This limits the possibility to use CsI-based gaseous PDs in harsh environment. The main source of ion bombardment is the ion avalanche produced in the multiplication process; the fraction of ions reaching the cathode depends on the detector architecture.

In recent years, MPGD-based PDs with enhanced ion blocking capability have been developed [3] and implemented for physics measurements. The four MPGD-based PDs [4] of COMPASS RICH-1 represent the most successful application of this technology: they have a hybrid architecture consisting of two THGEM multiplication layers (the first one coated with CsI) and a Micromegas. They cover a total active area of 1.4 m² and are in operation since 2016.

The development of more robust photoconverters for gaseous PDs, for future RICH applications at new facilities, for the timing resolution frontier, etc. needs a dedicated effort.

The EPIC collaboration [5] at the EIC [6], a facility aimed to achieve understanding of Quantum ChromoDynamics (QCD), in particular in the elusive non-perturbative domain, requires efficient hadron identification in a wide particle momentum range, imposing the use of the gaseous RICH technique. The compact design of the EPIC Detector imposes limitations on the RICH radiator length. A possible solution could be the detection of photons in the very far UV range by windowless gaseous PDs operating with the radiator gas itself [7], with high gain at high rate. This solution challenges the CsI performance limitations.

The quest for an alternative UV-sensitive photocathode overcoming these limitations is the main motivation for the R&D programme discussed in this article. We first recall the novel procedure to produce Hydrogenated NanoDiamond (H-ND) based photocathodes and the recent results on the photon extraction efficiency in different Ar/CH₄ gas mixtures. We then

*Corresponding author

Email address: fulvio.tessarotto@ts.infn.it (F. Tessarotto)

illustrate the study of the compatibility between H-ND and THGEMs and present first results from the systematic comparison of the response of THGEMs as electron multipliers before and after the H-ND coating. All THGEMs used for the studies reported in this article have an active area of $30 \times 30 \text{ mm}^2$ and the same geometrical structure as COMPASS RICH-1 THGEMs: hole diameter of 0.4 mm, pitch of 0.8 mm, thickness of $470 \mu\text{m}$ and no rim around the holes.

2. Nanodiamond-based photocathodes preparation and first trials on THGEMs

The high QE value of CsI photocathodes is related to its low electron affinity (0.1 eV) and wide band gap (6.2 eV) [8]. The NanoDiamond (ND) particles have a comparable band gap of 5.5 eV and low electron affinity of 0.35-0.50 eV, and they exhibit chemical inertness and good radiation hardness: they represent a potential candidate for photo-converter material more robust than CsI. ND hydrogenation lowers the electron affinity to -1.27 eV, favoring an efficient escape into vacuum of the generated photo electrons [9].

The standard procedure of hydrogenation of ND powder photocathodes is performed by using the MicroWave Plasma Enhanced Chemical Vapor Deposition (MWPECVD) technique at $\sim 800 \text{ }^\circ\text{C}$. The hydrogenated powder is then cooled down to room temperature under high vacuum. This procedure cannot be used for THGEMs which are made of fiberglass, and do not tolerate temperatures above $180 \text{ }^\circ\text{C}$.

A novel coating procedure, developed in Bari [11, 12], provides high QE [9] and low-temperature ($\sim 120 \text{ }^\circ\text{C}$) deposition, allowing to cover different types of substrates, including standard THGEMs, with H-ND photoconverting layers.

The ND or H-ND powder is mixed with deionized water in one to one proportion and sonicated for 60 minutes. The solution is sprayed using a pressure atomizer with a LabVIEW software controlled system in pulses (shots) of 100 ms at about 3 Hz frequency.

The substrates are kept at $\sim 150 \text{ }^\circ\text{C}$ and rotated by magnetic stirrer; the distance between the atomizer nozzle and the substrate is $\sim 10 \text{ mm}$. We now deposit $\sim 250 \text{ shots/cm}^2$, according to the results of a previous study of photoemission as function of the number of shots, in which we observed that the photocurrent saturated above 50 shots/cm^2 , as can be seen in Fig. 1.

After the coating a heat treatment (18 h in oven at $120 \text{ }^\circ\text{C}$) is applied: this treatment has been verified not to alter the QE response of the samples.

Preliminary measurements of the photosensitivity of PCB substrates coated with ND and H-ND have been performed before fixing the protocol described above: while the standard PCB substrate samples were always offering promising results, some of the first coated THGEM samples could not be operated as electron multipliers due to severe losses in the electric strength after being coated with H-ND.

A systematic study was then started to verify the compatibility of H-ND with THGEMs: a precise definition of the production and test procedures and an accurate characterization of

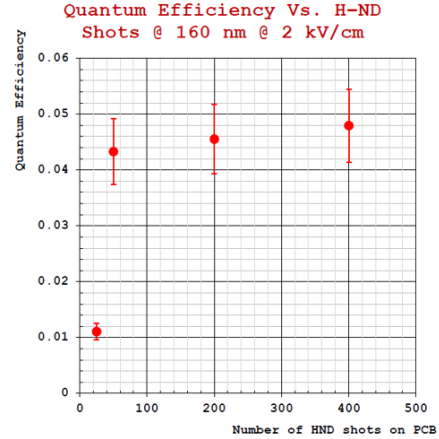


Figure 1: The QE of H-ND coated PCB substrates for 160 nm photons has been measured for samples coated with different quantity of H-ND powder, in units called shots. For amounts corresponding to 50 shots or more the QE is constant, indicating sufficient surface coverage.

the response of many THGEM samples before and after coating are ongoing. In parallel, a test of the aging properties of H-ND coated samples [14] was performed at CERN and showed that H-ND is at least ten times more robust than CsI against ion bombardment.

3. Measurement of H-ND photocurrent in different gas mixtures

Three types of ND powders have been used in 2021 for the new study of both QE and THGEM compatibility. Two types have an average grain size of 250 nm and were produced by Diamonds & Tools (D&T) srl and Element 6 (E6); the third type, boron doped (BDD) produced by SOME BETTER : ChangSha 3 Better Ultra-hard materials Co.,LTD has an average grain size of 500 nm.

Test substrates have been coated with the three types of powders, both without hydrogenation and with hydrogenation, and their QE was measured for each of them, for different wavelengths, and different applied electric field values: the results of this systematic study are reported in another article [15]; the hydrogenated D&T ND powder provides the highest quantum efficiency for all wavelengths in vacuum.

Systematic measurements were also performed at the peak wavelength of 160 nm for different electric fields in different Ar/CH₄ gas mixtures. We present in Fig. 2 the measurement of photocurrent as function of applied electric field for seven gas mixtures. For electric field value lower than 1 kV/cm, the measured photo current in argon rich gas mixtures is lower compared to that of CH₄ rich mixtures, due to the high photon back scattering cross section of argon.

For mixtures with low percentage of CH₄, the effect of gas multiplication is visible for electric field values larger than 0.5 kV/cm; above $\sim 1.5 \text{ kV/cm}$ the mixtures rich in argon provide larger total measured photocurrent values compared to CH₄ rich ones, because the gas multiplication effect becomes dominant.

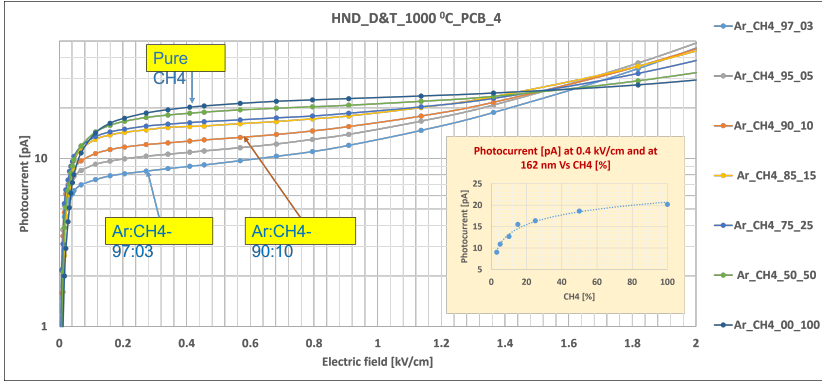


Figure 2: Photocurrent as a function of applied electric field over the photocathode surface for different Ar/CH₄ gas mixtures at 160 nm wavelength. The photocurrent values for 0.4 kV/cm as function of the fraction of CH₄ are presented in the inserted colored canvas.

The photocurrent values for 0.4 kV/cm as function of the fraction of CH₄ are presented in the colored canvas inserted in Fig2: With respect to the pure CH₄ gas case, the Ar/CH₄ 50/50 mixture shows a 9% lower photocurrent value, the Ar/CH₄ 75/25 a 24% lower value and the Ar/CH₄ 97/03 a 55% lower value.

4. Study of H-ND compatibility with THGEMs

The characterization of each THGEM is performed in a simple test setup including a plane of drift wires above the THGEM and a segmented readout anode plane below it, both properly biased, providing the drift and induction field respectively. The detector is operated with various gas mixtures, all including argon. The electrons from ⁵⁵Fe X-Rays, converted by argon, are collected and multiplied in the THGEM holes. The electron avalanche generated in the multiplication process, while drifting towards the anode, induces the detected signal.

All THGEMs have been characterized using a Ar/CO₂ 70/30 gas mixture and a ⁵⁵Fe X-ray source at INFN Trieste before applying the spray procedures, with the goal of performing comparative studies after coating them with VUV sensitive films. Environmental conditions (gas pressure and temperature) were registered and used for the corrected effective gain determination. The THGEM gain response has a variation in time related to the effect of charging up of the dielectric surfaces: a gain evolution study of about three days has been performed for each THGEM sample. The same characterization has been repeated after coating.

A typical ⁵⁵Fe X-ray spectrum obtained in Ar/CO₂ 70%/30% gas mixture is shown in the top panel of Fig. 3. The bottom panel shows the gain dependence of the THGEM as function of the applied bias voltage.

Each THGEM sample has been coated in Bari either with as-received ND powder, or with H-ND. To allow easier comparison, for some THGEMs only half of the active surface has been coated, leaving the other half uncoated. The same coating procedure (and the same amount of ND per unit surface) has been applied to the PCB test substrates and to the THGEMs.

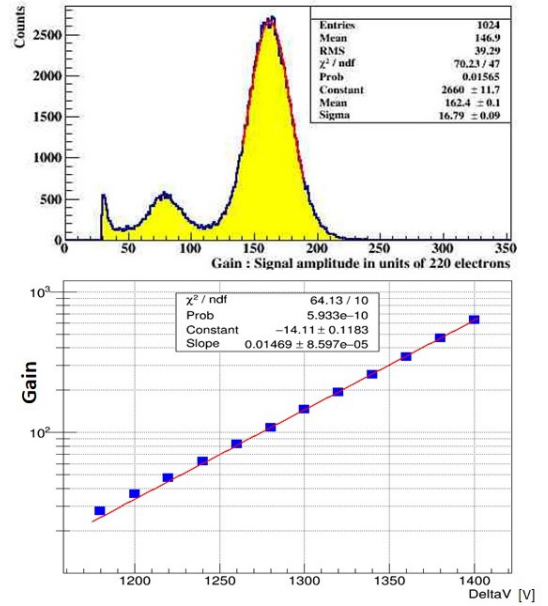


Figure 3: Top: typical ⁵⁵Fe X-ray spectrum obtained in Ar/CO₂ 70%/30% gas mixture when the applied voltages at drift, top and bottom of THGEM are -2520 V, -1720 V and -500 V, respectively, while the anode is at ground voltage. Bottom: gain dependence of the THGEM as function of the applied voltage.

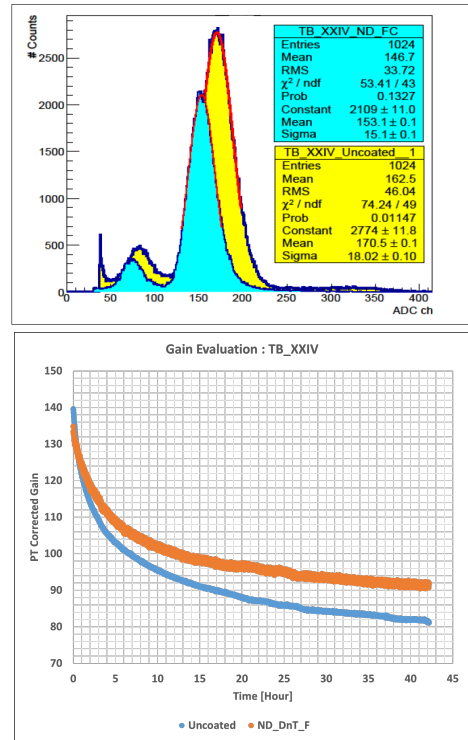


Figure 4: Top: typical ⁵⁵Fe X-ray spectra obtained from a same THGEM-XXIV before coating and after fully coated ND powder in Ar/CO₂ 70%/30% gas mixture. The voltages applied to drift, top and bottom of the THGEM electrodes are -2250 V, -1750 V and -500 V respectively; the anode is kept at ground voltage. Bottom: gain evolution due to charging-up of the THGEM measured before (blue) and after (orange) coating; corrections for pressure and temperature have been applied.

Some of the fully characterized THGEMs and their respective coatings are listed below:

Uncoated THGEMs	Max ΔV	Coated THGEMs	Max ΔV
TB_XV	1325	H-ND_BDD_H	1325
TB_XVI	1375	ND_E6_H	1400
TB_XVII	1350	ND_BDD_H	1325
TB_XXIV	1375	ND_D&T_F	1325

Table 1: Maximum stable voltage of few uncoated and coated THGEMs in Ar/CO₂ 70%/30% gas mixture.

The first column of Tab.1 shows the THGEM labels, the second one the maximum voltage bias at which the THGEM was operating stably; in the third column the coating material for each THGEM are listed and in the fourth the maximum stable voltage after coating is provided. The electrical stability of THGEMs does not seem to be systematically affected by the coating layers. The variations are compatible with the changes in environmental conditions.

Typical examples of amplitude distributions for ⁵⁵Fe signals from an uncoated (yellow) and ND coated (light blue) THGEM are shown superimposed in the top part of Fig. 4. The amplitude spectra are presented before application of the correction for the different pressure and temperature conditions. The effective gain is extracted from the mean of the Gaussian fit of the main peak of the spectrum. To study the charging-up response, a continuous set of short (~ 30 sec.) acquisition runs over few days is performed. In the bottom part of Fig. 4 the charging-up curve is presented for the same THGEM before (blue) and after (orange) coating; for this comparison the effective gain values have been corrected for pressure and temperature. The difference seen in the charging up variation rate is partly due to the change in the ⁵⁵Fe source activity.

A typical plot of the effective gain values, corrected for pressure and temperature, as a function of the bias voltage applied to the THGEMs is presented in Fig. 5. The blue points represent the corrected gain values before coating; the red ones the gain values after coating. The characterization of a THGEM without and with various ND coatings provides almost identical results. The gain values are found to be similar in all cases: the THGEM response as electron multiplier is not significantly modified by any of the ND and H-ND coatings.

To achieve a more accurate evaluation of possible small effects, more THGEM samples will be tested. A newly built, complete photon detector prototype is now available and will be used in a campaign of tests for the next step in the validation of the "MPGD + H-ND technology" for the detection of single VUV photons for RICH applications. Other fields can potentially benefit from the development of this innovative technique.

5. Conclusion

The ongoing R&D programme to explore the H-ND photocathode response in the VUV range for future gaseous PD application has provided encouraging results. Photoemission

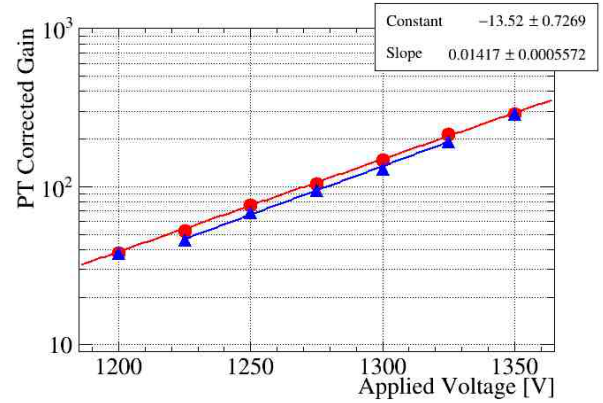


Figure 5: Comparison between corrected effective gain versus applied voltage across the THGEM electrodes before (blue) points and after (red points) coating.

current measurements in the VUV range in vacuum and various gas mixtures were performed for ND and H-ND samples. Photoemission currents measured at 160 nm as a function of the electric field, in various Ar/CH₄ compositions, allowed to distinguish between two different voltage regimes. At lower field values the important role of CH₄ in reducing the photoelectron backscattering is highlighted, providing a factor of two in photoconversion probability between Ar-rich mixtures and pure CH₄. At higher voltages the gas multiplication dominates, in particular for the gas mixtures with large argon fraction.

THGEM samples coated with different types of photosensitive layers of ND and H-ND, from D&T, E6 and BDD have been studied with extensive characterization. The studied THGEMs sustain more than 1325 V bias across top and bottom surfaces. The response of THGEMs as electron multipliers is not significantly modified by the ND or H-ND coating. The results of this first exploratory study, suggesting a full compatibility of H-ND with THGEMs encourage to proceed in the development of gaseous PDs with a photocathode overcoming the limitations of CsI.

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References

- [1] Triloki n, B.Dutta,B.K.Singh, *Influence of humidity on the photoemission properties and surface morphology of cesium iodide photocathode*, Nucl. Instr. and Meth. A, **695** (2012) 279.

- [2] H.Hoedlmoser et al., *Long term performance and ageing of CsI photocathodes for the ALICE/HMPID detector*, Nucl. Instr. and Meth. A, **574** (2007) 28.
- [3] A. Bondar et al., *Study of ion feedback in multi-GEM structures*, Nucl. Instr. and Meth. A **496** (2003) 325;
 A. Breskin et al., *Sealed GEM photomultiplier with a CsI photocathode: ion feedback and ageing*, Nucl. Instr. and Meth. A **478** (2002) 225;
 J.F.C.A. Veloso et al., *A proposed new microstructure for gas radiation detectors: The microhole and strip plate*, Rev. Sc. Instr. **71** (2000) 2371;
 A.V. Lyashenko et al., *Further progress in ion back-flow reduction with patterned gaseous hole - multipliers*, JINST **2** (2007) P08004;
 A.V. Lyashenko et al., *Efficient ion blocking in gaseous detectors and its application to gas-avalanche photomultipliers sensitive in the visible-light range*, Nucl. Instr. and Meth. A **598** (2009) 116;
 M. Alexeev et al., *Ion backflow in thick GEM-based detectors of single photons*, JINST **8** (2013) P01021.
 M.M. Aggarwal et al, (ALICE TPC Collaboration), *Particle identification studies with a full-size 4-GEM prototype for the ALICE TPC upgrade*, Nucl. Instr. and Meth. A **903** (2018) 215.
- [4] J. Agarwala et al., *The MPGD-based photon detectors for the upgrade of COMPASS RICH-1 and beyond*, Nucl. Instr. and Meth. A **936** (2019) 416.
- [5] R. Abdul Khalek et al., *Science Requirements and Detector Concepts for the Electron-Ion Collider: EIC Yellow Report*, Nuclear Physics A **1026**, (2022), 122447
- [6] A. Accardi et al., *Electron-Ion Collider: The next QCD frontier*, Eur. Phys. J. A **52** (2016) 268.
- [7] M. Blatnik et al., *Performance of a Quintuple-GEM Based RICH Detector Prototype*, IEEE NS **62** (2015) 3256.
- [8] A.Buzulutskov, A.Breskin, R.Chechik, *Field enhancement of the photoelectric and secondary electron emission from CsI*, Journal of Applied Physics **77** (1995) 2138.
- [9] L. Velardi, A. Valentini, G. Cicala, *Highly efficient and stable UV photocathode based on nanodiamond particles*, Appl. Phys. Lett. **108** (2016) 083503-1-5.
- [10] H. Rabus, et. al., *Quantum efficiency of cesium iodide photocathodes in the 120-220 nm spectral range traceable to a primary detector standard*, Instr. and Meth. A , **438** (1999) 94.
- [11] G. Cicala, A. Massaro, L. Velardi, G. S. Senesi, A. Valentini, *Self-Assembled Pillar-Like Structures in Nanodiamond Layers by Pulsed Spray Technique*, ACS Appl. Mater. Interfaces **6** (2014) 21101.
- [12] L. Velardi, A. Valentini, G. Cicala, *UV photocathodes based on nanodiamond particles: effect of carbon hybridization on the efficiency*, Diam. Relat. Mater. **76** (2017) 1-8.
- [13] A. Valentini, D. Melisi, G. De Pascali, G. Cicala, L. Velardi, A. Massaro, *High-efficiency nanodiamond-based ultraviolet photocathodes*, 30-03-2017 Patent n. **WO 2017/051318 A9**; International Patent n. **PCT/IB2016/055616** of September 21, 2016; National Patent Italia - n. 102015000053374 del 21 Settembre 2015, Istituto Nazionale di Fisica Nucleare e Consiglio Nazionale delle Ricerche.
- [14] F.M. Brunbauer et al., *Employment of nanodiamond photocathodes on MPGD-based HEP detector at the future EIC*, J. Phys.: Conf. Ser. **2374** 012140.
- [15] F.M. Brunbauer et al., *Study of nanodiamond photocathodes for MPGD-based detectors of single photons*, submitted to Nucl. Instr. and Meth. A.