Available online at www.sciencedirect.com
SCIENCE DIRECT®
ELSEVIER
Nuclear Instruments and Methods in Physics Research A 554 (2005) 220–225
NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH Section A
www.elsevier.com/locate/nima

Ablated Mg films with a graphite cover as photocathodes

A. Perrone[a], L. Cultrera[a], A. Pereira[b], M. Rossi[c], S. Cialdi[d], I. Boscolo[d], F. Tazzioli[e,*], C. Vicario[e], G. Gatti[e]

[a]*Physics Department and INFN, University of Lecce, Via per Arnesano, 73100-Lecce, Italy*
[b]*LP3, CNRS/University of Mediterranee, 163 Avenue de Luminy, C917, 13288 Marseille, France*
[c]*Dipartimento di Energetica, University of Roma "La Sapienza", Via A. Scarpa 16, 00161 Roma, Italy*
[d]*University and INFN, Via Celoria 16, 20133 Milano, Italy*
[e]*INFN-LNF Via E. Fermi, 40, 00044 Frascati-Roma, Italy*

Received 11 July 2005; accepted 16 August 2005
Available online 15 September 2005

Abstract

Mg films deposited by pulsed laser ablation on a Cu substrate were tested as photocathodes in a DC diode. They showed features of robustness and a quantum efficiency of 3×10^{-4} at low accelerating field when illuminated by 266 nm light at normal incidence. A graphite protective layer of about 20 nm thickness deposited by laser ablation just after the Mg deposition made the cathode stable and of easy handling. The cathodes could stay for several weeks in air before being inserted in the test chamber. The preparation procedure and the tests are presented.

PACS: 29.25.Bx; 79.60.Dp; 81.15.Fg

Keywords: Thin films; Photocathodes; Laser ablation

1. Introduction

The search for a rugged and efficient photocathode for advanced high-brilliance electron injectors, after a decade of worldwide efforts is still in order. Most new designs of such injectors envisage an electron source consisting of a Radio frequency gun (RF gun) with a photocathode excited by an UV laser pulse. The preferred wavelength is 266 nm, that is the third harmonic of Ti:Sa laser. Such lasers can provide the very short, sub-picosecond rise time, photon pulses required by the injectors [1]. That wavelength corresponds also to the fourth harmonic of the Nd:Yag laser that we have used for our quantum efficiency (QE) measurements. The present work is concerned with the development of rugged photocathodes for such injectors. The paper presents a photocathode based on a Mg film deposited by

*Corresponding author. Fax: +39 06 9403 2565.
E-mail address: franco.tazzioli@lnf.infn.it (F. Tazzioli).

0168-9002/$ - see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2005.08.078

pulsed laser ablation deposition (PLAD) technique and covered by a graphite protective film. The proposed solution relies on the availability of laser beams with so high-energy density as to consent ablating the upper layer of the cathode surface for activation. Progress in cesiated materials, especially alkali-tellurides, has, by now, resulted in QE of few percent, at 266 nm laser wavelength, and life times of weeks [2,3]. However, such cathodes require ultrahigh vacuum environment, being highly reactive; moreover, they have rather slow time response. Metal photocathodes are inherently robust but they have a low quantum efficiency even with ultraviolet (UV) radiation, thus they require high-power UV lasers for cathode irradiation. Magnesium has been identified, since long [4,5], as a metal photoemitting material with relatively high QE, ~0.3% at 266 nm wavelength [6]. The features of efficiency, robustness, promptness and good emittance of Mg disks have been investigated with a certain success within RF-guns [7,8]. Mg disks installed in RF-gun with the press fitting technique had problems of RF breakdown at the joint between the Mg disk border and the Cu embedding structure. Frictional welding technique did not solve completely the breakdown problems [9]. Moreover, the distribution of QE shows large variations over the spot irradiated by the laser [10]. This nonuniform distribution causes a deterioration of the brilliance of the electron source [11]. Mg film deposited on metal disks by sputtering also showed problems of RF breakdown during the RF conditioning. This is guessed to be due to insufficient smoothness of the cathode surface and bad adherence to the substrate. Anyway, cathodes made up by thin films are expected to present a better emission uniformity and a lower discontinuity at the border. With the aim of obtaining tightly bounded films with very smooth surfaces, so to overcome those problems, we prepared Mg photocathodes with the PLAD. In this deposition technique the ablated material particles have high energy, order of 50 eV in our experiments, against 10 eV of magnetron sputtering. Thus, the nucleation process at the surface is improved compared with other deposition processes [12]. The PLAD Mg films used in our tests were about 0.5 μm thick and covered with a 20 nm graphite layer for protection purpose, because of the high reaction rate of Mg with oxygen and other gases in the atmosphere. Photocathode protection with thin films, both permanent or removable, has already been experienced [13,14]. In our case, the novelty is in the use of a graphite protective layer on a Mg film. The permanent solution is not viable because of the low electron transmission of graphite. The protective layer is removed by laser ablation prior to the photocathode use. This cathode arrangement made the cathode of safe maintenance and easy handling [15]. Protection with a graphite film is more efficient than with an MgO film because oxidation can penetrate deeply within the Mg crystal and this thick layer can be hard to remove [16]. The tests on PLAD grown Mg films as photocathodes presented here are the first as far as we know.

2. Sample preparation

The films were obtained by deposition of material ablated by a pulsed laser beam of 308 nm wavelength (Lambda Physics LPX 315i *XeCl* excimer laser providing pulses of $\tau = 30$ ns). In Fig. 1 we show a sketch of the sample preparation technique. The target where the laser beam was focused rotated at a frequency of 1 Hz. The impinging angle of the light beam with respect to the target surface was 45°. The target was arranged as a disk with a 12 mm diameter graphite

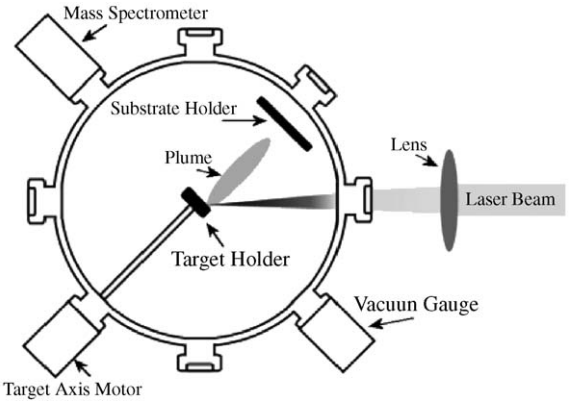

Fig. 1. Sketch of film preparation process.

inner core fitted within a thick ring of high-purity Mg (25 mm external diameter). This special target configuration allowed us to perform the deposition of the Mg film and in succession (within a unique process) of the protective graphite film coating. In fact, the laser beam was first launched onto the Mg external ring and in succession onto the graphite core by a simple shift of the laser beam on the target. Several depositions were performed at different laser parameters inside a high-vacuum (HV) chamber set at 5×10^{-6} Pa pressure, with the aim of obtaining thick films, well adherent to the substrate. The quality of the vacuum was controlled by a quadrupole mass spectrometer before, after and during depositions. We have singled out for discussion two of these films (samples 1 and 2) that have shown better uniformities. The experimental parameters used for such depositions are reported in Table 1.

The characteristics of the deposited film depend on the ablation rate, the plume deflection and the morphological changes of target surface created by prolonged laser irradiation. The right set of parameters for a thick and adherent film has been chosen via systematic tests on the results obtained with different apparatus arrangements [16]. In order to deposit one Mg film, 30.000 subsequent laser pulses were applied. Then 2.000 and 3000 laser pulses were applied to deposit a thin layer of graphite, respectively, for samples 1 and 2 . Before each successive deposition run, the target surfaces were cleaned by applying a series of laser pulses as shown in the table. During these cleaning processes, a shutter was interposed between the target and the Cu substrate.

Table 1
Experimental conditions used for the Mg thin film deposition

Sample	1	2
Target	Mg-C	Mg-C
Substrate	Cu	Cu
Target–substrate distance	4.5 cm	4.5 cm
Laser spot size	0.90 mm^2	0.96 mm^2
Base pressure	5×10^{-8} mbar	5×10^{-8} mbar
Laser pulses		
Mg cleaning	5000	5000
Mg deposition	30 000	30 000
C cleaning	1000	2000
C deposition	2000	3000
Laser fluence	6 J/cm^2	10 J/cm^2
Quantum efficiency	5×10^{-5}	3×10^{-4}

3. Experimental setup, results and discussion

The samples were tested as photocathodes in a vacuum photodiode cell at 10^{-6} Pa and with an applied electric field of 1 MV/m. The Mg cathode was irradiated by a frequency quadrupled (266 nm, 30 ps) Nd:YAG laser, at 0° incidence angle. For removing the protective graphite layer, we used the so-called laser cleaning technique. A succession of trains of several hundred laser pulses is applied and the QE is periodically monitored after each train. The laser spot size had a diameter of 0.3 mm with a power density of about 1 GW/cm^2 (that is 300 $\mu J/mm^2$). The laser beam swept a circular area up to 2 mm in diameter. Then the laser spot size for cathode excitation was set at the diameter of the cleaned area. The QE measurements were made with much lower power density to avoid space charge saturation and laser ablation processes. Fig. 2 shows typical emission curves before and after laser cleaning. The QEs of samples 1 and

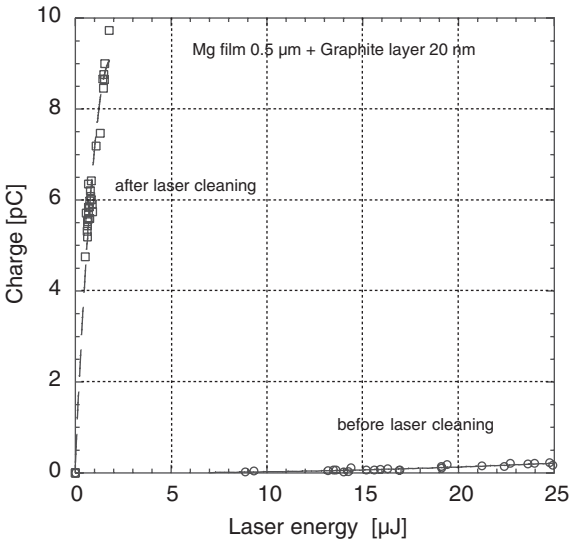

Fig. 2. Intensity dependence of the yield at 266 nm and normal incidence for sample 1, before and after laser cleaning.

2 ranged in the interval 5×10^{-5}–3×10^{-4}. This large QE spread could be attributed both to different film features and to a not yet optimized and reproducible laser cleaning process. Our tests were performed after storing the samples in air for several weeks. This indicates that the protective graphite layer is effective on the long term.

The EDX analysis of the composition distribution showed that the graphite protective layer was not completely removed, scattered graphite patches were still on the surface. A diffused presence of oxygen was also observed. The first layers of the films were certainly oxidized when extracted from the vacuum environment for being analyzed, but we may also argue that some MgO molecules could have been formed during the deposition process. Moreover, the SEM analysis revealed that the laser irradiation had caused an uneven ablation of the film surface. In fact, the Mg film resulted nearly completely removed at few tiny spots. This indicates that the laser power density for surface cleaning must be less than $300\,\mu J/mm^2$. We would also add that the surface of the films did not result flat and uniform at the best. Atomic force microscope analysis could not be well performed because the roughness of the substrate was comparable with the film thickness. The SEM analysis instead has given significant results and confirmed that the film structure is amorphous. The morphological features are substantially different from those characterizing the sputtered Mg films that we have analyzed for comparison, as discussed further on. Those sputtered films presented a well-oriented columnar crystalline morphology. In Fig. 3 we show a microscopic SEM image of the surface of the PLAD film before and after laser cleaning. The analyzed zone has been chosen at first by visual inspection, to make sure that the red color of Cu did not appear, thus indicating that the film had not been punched. Then, when examined at the SEM microscope, the irradiated zone, cleaned of graphite, was clearly distinguishable as much brighter than the non-irradiated one. This happens because MgO has a much higher secondary emission than graphite.

To compare, in the same experimental conditions, our Mg PLAD films with sputtered Mg films already applied as photocathodes for injectors in other laboratories, we have tested some magnetron sputtered films produced by industry. These films were 10 μm thick and had been transported to us under nitrogen atmosphere. An oxide layer, that can penetrate as deep as 150 nm, forms naturally when the film is exposed to air after the deposition. In our case the exposition to air during installation was about 20 min. The maximum QE we measured, after ablation of the oxide layer, was 10^{-4}. Our PLAD films are covered with a thin graphite layer under vacuum, immediately after the Mg deposition, in order to protect the Mg from deep oxidation. This is important because PLAD films cannot be grown much thicker than few microns. The film is extracted from the deposition chamber already covered with graphite. The measured

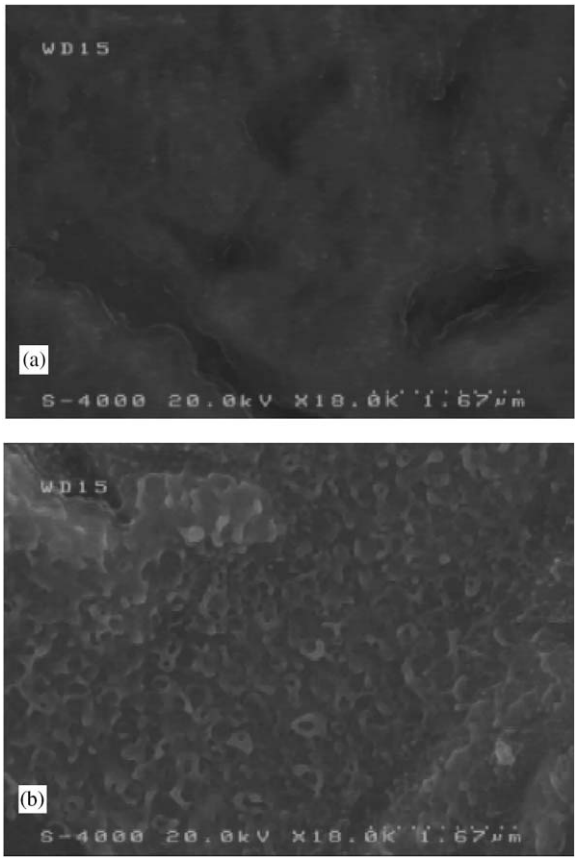

Fig. 3. Microscopic image of the surface of the film, without and with irradiation.

photo-emission efficiency before graphite removal is that of graphite itself, that is, extremely low at 266 nm wavelength. This shows that there is no transmission of either photons or electrons across the graphite layer. After removal of the graphite layer we have measured a maximum QE of 3×10^{-4}, that is already interesting for the application. A previous test made on thin films (about 200 nm) without the graphite protection, confirmed that the film had been strongly oxidized and the QE was therefore very low (in the 10^{-6} range). Further tests to improve PLAD film uniformity and thickness have been already planned. It remains to be explained why, for both the PLAD and sputtered films that we tested, the QE at low field (1 MV/m) was lower than the maximum value (0.3%) of sputtered Mg films reported in the literature [6]. In our prototype PLAD Mg films, the difference could be due to residual graphite clusters and the observed absence of Mg in various zones within the cleaned area. In the sputtered films the difference could be attributed to incomplete removal of the oxide layer. Table 1 shows a correlation between laser fluence and QE, but this difference could also be due to not yet reproducible laser cleaning operations both during film deposition and for cathode activation before QE measurements. The film compactness and adhesion to the substrate could be enhanced by increasing the laser fluence because of the higher kinetic energy of the ablated material. This should also have a beneficial effect on QE due to the increase of the useful emitting area. We intend to explore the effect of the laser fluence up to tens of J/cm^2 and to improve the film purity by lowering further the base pressure. In addition, we intend to set up a well controlled and gradual laser cleaning process, both during the deposition process and for cathode activation. These improvements require an up-graded system that is in course of preparation.

In conclusion, Mg thin films deposited by PLAD technique have shown very good QE, up to 3×10^{-4} at low accelerating field and in a not extreme vacuum environment. A 20 nm graphite film resulted in a completely efficient protective film. It is a simple technique for conserving spare photocathodes within a laboratory and it allows an easy handling of them. Advanced electron injectors require an emitted charge of about 1 nC from a 2 mm diameter spot. If the QE is in the order of 10^{-4}, the required laser energy density is much lower than the experienced damage threshold of $300\,\mu J/mm^2$. The challenges still confronting us are to obtain a thick and uniform film and to devise a non-destructive surface cleaning technique for the activation of the cathode.

Acknowledgements

We express our hearty thanks to Dr Silvia Orlanducci of Rome2 University, Dipartimento di Scienze e Tecnologie Chimiche, for the accurate SEM analysis of our samples.

The work in this article is partly supported by Ministero Istruzione Universita' Ricerca, *Progetti Strategici*, DD 1834, Dec. 4, 2002 and European Contract RII3-CT-PHI506395CARE.

References

[1] Technical design report for the SPARC advanced photo-injector, ⟨http://www.lnf.infn.it/acceleratori/sparc/⟩.
[2] I. Boscolo, P. Michelato, Nucl. Instr. and Meth. A 445 (2000) 389.
[3] G. Suberlucq, Proceedings of the EPAC Conference, 2004, p. 64.
[4] T. Srinivasan-Rao, J. Fisher, T. Tsang, J. Appl. Phys. 77 (1995) 1275.
[5] T. Srinivasan-Rao, D.T. Palmer, I. Ben-Zvi, R.H. Miller, J. Smedley, X.J. Wang, M. Woodle, Proceedings of the IEEE Particle Accelerator Conference, vol. 3, 1997, p. 2790.
[6] T. Srinivasan-Rao, J. Schill, I. Ben Zvi, M. Woodle, Rev. Sci. Instr. 69 (1998) 2292.
[7] X.J. Wang, M. Babzien, R. Malone, Z. Wu, Proceedings of the LINAC Conference, 2002.
[8] X.J. Wang, T.S. Rao, K. Batchelor, I. Ben-Zvi, J. Fischer, Nucl. Instr. and Meth. A 356 (1995) 159.
[9] X.J. Wang, M. Babzien, R. Malone, J. Sheehan, J. Skaritka T. Srinivasan-Rao, M. Woodle, V. Yakimenko L.H. Yu, Proceedings of the IX Linac Conference Chicago, 1998, p. 866.
[10] D.T. Palmer, The next generation photoinjector, Ph.D. Thesis, Stanford University, Department of Applied Physics, UMI-99-01569-mc, 1998.
[11] M.Quattromini, L.Giannessi, C.Ronsivalle, Spectral analysis of charge emission spatial inhomogeneities, FEL Conference, Trieste, 2004.

[12] A.A. Voevodin, M.S. Donley, Surf. Coating Technol. 82 (1996) 199.
[13] A. Breskin, et al., Nucl. Instr. and Meth. A 413 (1998) 275.
[14] A. Buzulutskov, et al., Nucl. INstr. and Meth. A 400 (1997) 173.
[15] Q. Yuan, A.W. Baum, R.F.W. Pease, P. Pianetta, J. Vac. Sci. Technol. B 21 (2003) 2830.
[16] L. Cultrera, A. Pereira, C. Ristoscu, A. Clozza, F. Tazzioli, C. Vicario, Appl. Surf. Sci. 248 (2005) 397.