PREPARATION OF Pb-PHOTOCATHODES AT NATIONAL CENTRE FOR NUCLEAR RESEARCH IN POLAND - STATE OF THE ART*

J. Lorkiewicz [#] , I. Cieślik, P. J. Czuma, A. M. Kosińska. R. Nietubyć, National Centre for Nuclear Research (NCBJ), 05-400 Otwock-Świerk, Poland
J. Sekutowicz, Deutsches Elektronen Synchrotron (DESY), 22607 Hamburg, Germany

Abstract

R&D activities related to preparation of the superconducting Pb photocathode layer on niobium substrate are ongoing at the National Centre for Nuclear Research (NCBJ) in cooperation with DESY, HZDR, HZB, BNL and other research institutes. The activities are part of the R&D program at DESY for the cw-upgrade of E-XFEL and for the newly approved free electron laser facility PolFEL to be built and operated at NCBJ. The optimization results obtained for the lead deposition on niobium and smoothing of the coated layers are reported. The photocathodes samples were tested for their surface morphology, microstructure and quantum efficiency in terms of the impact on the operation of all-superconducting RF electron injector, proposed for both facilities.

INTRODUCTION

Research and development program on a 1 mA-class, fully-superconducting radio-frequency (SRF) electron photo-injector is an integral part of the task of improving the duty factor of X-ray free electron laser (XFEL) at DESY [1-3] and in other, similar devices. The injector is expected to produce electron beam bunches of normalized emittance below 1 µrad with charges up to nC. Long pulse and continuous wave operation modes are also anticipated [1]. The concept of a superconducting RF, hybrid Nb-Pb electron injector for linear superconducting (sc) accelerators was proposed and developed [1,4-8]. This solution assumes the use of a fully sc photocathode in the form of a Pb film applied to niobium surface. Whereas both metals are superconductors with similar critical temperatures, lead exhibits much higher quantum efficiency (QE). Besides, a much shorter time of Cooper pairs recombination in Pb (a few µs) than in Nb at the 2 K working temperature [9] permits the exposure of such photocathode with laser beam at a repetition of up to 100 kHz.

Different design versions of the injector, comprising half-cell and 1.6-cell niobium cavities, based on TESLA SRF technology, were built, tested and optimized within a collaboration between DESY, Brookhaven National Laboratory (BNL), Stony Brook University, Thomas Jefferson National Accelerator Facility (TJNAF), NCBJ and Stanford Linear Accelerator Center (SLAC). To emit photoelectrons the lead photocathode is typically excited with an UV beam of wavelength between 193 nm and 266 nm, synchronized with the phase of RF field. The amplitude of the electric field on the cathode surface is 40-60 MV/m [9]. At this electric field the cathode is exposed to a magnetic field of up to 4 mT which is much smaller than the critical field of lead (80 mT). The optimized 1.6-cell injector structure was equipped with an exchangeable niobium "plug" mounted to the back cavity wall [10]. The lead photocathode is deposited on the plug tip. Such a solution, first proposed and tested at TJNAF [9], allowed avoiding the cathode contact with mixtures of acids used for chemical treatment of cavities. Nevertheless, high pressure water rinsing of the injector with the photocathode is needed to obtain sufficiently high field intensity in the cavity. This preparatory step imposes demands on the lead layer adhesion to the plug.

Usability of lead-layer hybrid photocathodes reached by different Pb coating procedures has been tested in a series of experiments. The deposition methods included: electroplating based on methane sulfonic acid chemistry [4], Pb evaporation in vacuum, magnetron sputtering, lead coating in cathodic arc [5] and by using pulsed laser deposition [11]. QE measurements were conducted in dedicated test-stands at a 1 MV/m dc extraction electric field at the cathode, after appropriate laser cleaning of its surface. The data presented in [4-5, 11] indicate that the Pb film coated in cathodic arc shows the highest QE. Its value (eg. QE=2.7x10^{-3} at a photon wavelength of 213 nm) is close to the theoretical predictions based on the Spicer three-step photoemission model [5, 12].

LEAD DEPOSITION IN CATHODIC ARC

Assuring proper and reproducible operation of a photocathode requires smooth and uniform lead film and its sufficient thickness. The film cleanliness, microstructure and morphology determine quantum efficiency, thermal emittance of photocurrent and dark current from the photocathode. These features, in turn, depend strongly on deposition and post-processing procedures used to prepare the layer.

Lead deposition in cathodic arc (also referred to as ultra-high vacuum (UHV) arc) is most promising due to the aforementioned high QE of electron photoemission from Pb layers obtained in this way. This type of arc is started and maintained without any support gas. It makes use of the eroded metallic cathode material as a discharge medium. However, the main disadvantage of using this coating method is the presence of Pb macroparticles (droplets), in a stream of plasma with lead atoms and ions. The droplets are by-products of activation of microscopic "cathode spots" on the arc cathode. They are sites of non-stationary,

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[#] email address: jerzy.lorkiewicz@ncbj.gov.pl

explosive emission of electrons and plasma generation. The occurrence of the macroparticles is linked with the impact of local plasma pressure on liquid metal present in these locations. In the absence of limitation in the flow of macroparticles between the arc cathode and the coated substrate, they are incorporated into the film which leads to dramatic increase of its roughness. This is particularly evident in the case of low-melting metals. The number of metallic droplets incorporated in a film decreases exponentially with their growing size [13]. Pb generate high number of droplets in the discharge. According to the literature data their sizes observed by scanning electron microscope (SEM) vary from 0.1 to 40 µm. Regardless of the arc current, the stream of lead macroparticles towards the substrate, normalized as the number within the size class of width 1 Å, per unit film thickness (in µm) and area (in mm^2) varies from ≈10^3 for particles sized at 0.5 µm down to ca. 0.001 for the spherical droplets 40 µm in diameter [14, 15].

The net effect of droplets incorporation into a 18 µm thick lead film, deposited in an arc without any droplets filtration, is shown in Fig. 1 with a Pb layer image obtained by using a scanning electron microscope (SEM, model: Zeiss EVO MA10). Numerous macroparticles of diameter from 0.5 up to 80 µm and of height up to 10 µm are visible. Smaller droplets sized up to a few µm assumed forms of hemispheres, larger – of donut-shaped craters. The latter resulted from collisions of big, liquid Pb drops with the target. The diameters of the craters are ca. twice as large as the diameters of drops from which they were created [13].

The photocathode surface roughness, in turn, enhances generation of dark current due to electron field emission during the gun operation. The dark current electrons are accelerated along with the photocurrent as an unwanted part of the beam which may result in beam loss, quenching of SRF structure, generating parasitic radiation doses which activate accelerator components or even in damaging an undulator structure [16-18]. It can also increase background for radiation beam users. For superconducting RF guns these parasitic effects cannot be ignored, particularly at continuous wave operation. On the other hand photo-emitter surface irregularities deteriorate beam quality by rising thermal beam emittance of photocurrent. Therefore, efforts have been made towards smoothing the Nb-Pb fotocathodes' emissive surface first by modifying the deposition system as such. To this end dedicated filters of various types were inserted between the cathode and the coated Nb substrate. Our initial approach included making use of passive filters - straight plasma ducts filled with mechanical baffles and chicanes which intercepted macroparticles while leaving some space for diffusion of arc plasma and enabling lead ions to reach the substrate (see [19] for details). Using these filters resulted in drop of the coating rate to 20 nm/min and to the presence in the coated layers of lead debris detached from the chicane surfaces. Therefore, the use of passive filters for preparing Nb-Pb photocathodes has been abandoned.

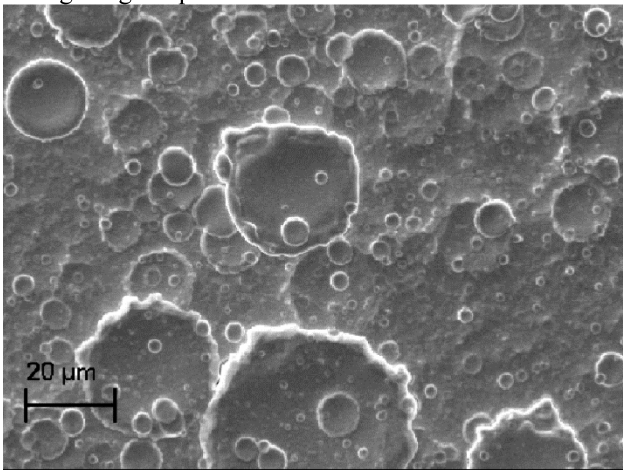

Figure 1: SEM image of a 18 µm thick Pb layer deposited without droplets filtering.

Alternatively, macroparticle reduction in a plasma stream can be accomplished in magnetic curved filters inserted between the cathode and substrate. In this case the arc ions are guided by generating magnetic field along a bended plasma duct [14]. In all the coating systems discussed below dc magnetic field of ≈10 mT was generated by external coils along the whole duct length. Unlike the plasma ions, macroparticles, due to their high mass and low charge, are not guided by the field but strike the curved duct wall where they are intercepted or reflected. Initial works at NCBJ with magnetically filtered arc were performed by coating lead films through a classic 90^0 duct filter (Aksenov-type [14]), directly on the back wall of a niobium gun resonator as reported in [8]. Using this tight filter resulted in reduction of Pb deposition rate down to 2 nm/min and in very small, non –uniform final film thickness (on average 0.1 µm) with visible niobium areas deprived of lead. After subjecting this Nb-Pb photocathode sample to laser cleaning the lead film suffered from melting and fragmentation [8].

To face the above problems with too thin and inhomgeneous Pb layer a less tight filter was used at NCBJ, bent by only 30^0. The whole plasma duct length amounted to 50 cm while the target was placed 10 cm downstream of the curved filter duct (Fig. 2). Unlike in the 90^0 duct filter, in this case the target could be negatively biased at 70 V in respect to the grounded chamber. It enhanced energetic deposition which resulted in homogenization of the film. The deposition rate has been increased by two orders of magnitude (up to 200 nm/min.) compared to Aksenov filter device. Therefore, the impurity content in the layer with oxygen and carbon has been reduced by a factor of 3. It has been checked with the energy dyspersive x-ray spectroscopy (EDS option of the used SEM) by using a 10 keV e$^-$ beam. The measurements revealed 2 and 1 weight % for C and O content , respectively, in the 2 µm thick Pb film deposited with the 30^0 - bent filter on a 1 x 1x 0.3 cm^3 planar Nb plate and on a special, cylindrical, 10 mm in diam. niobium plug (see Fig. 3). The reached basic layers were complete and smooth to within ±0.2 µm. On the other hand, due to dissipation on the walls of some solidified macro particles the 30^0 -bent filter proved not tight enough to prevent them from reaching the films. It

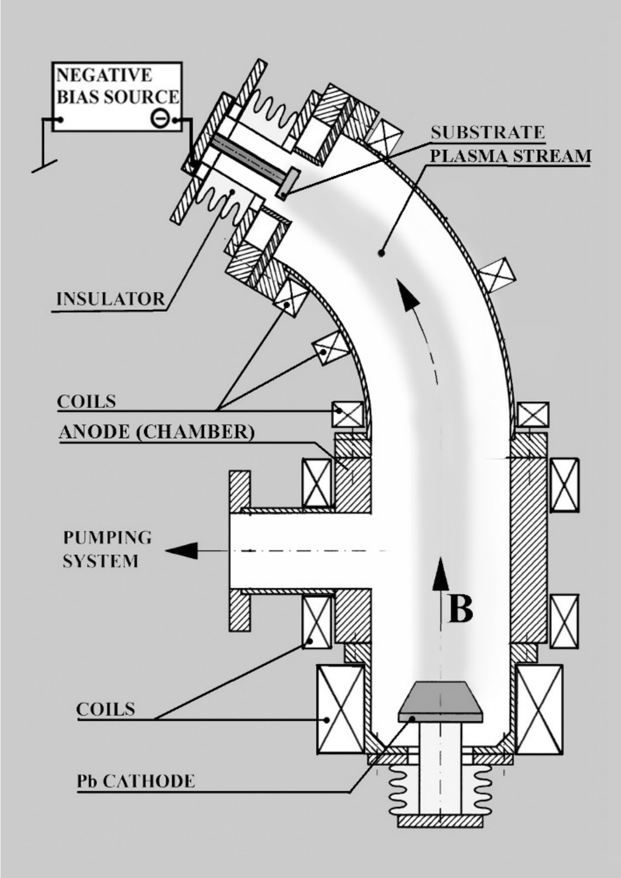

Figure 2: Coating device at NCBJ with cathodic arc and 30°- bent magnetic filter.

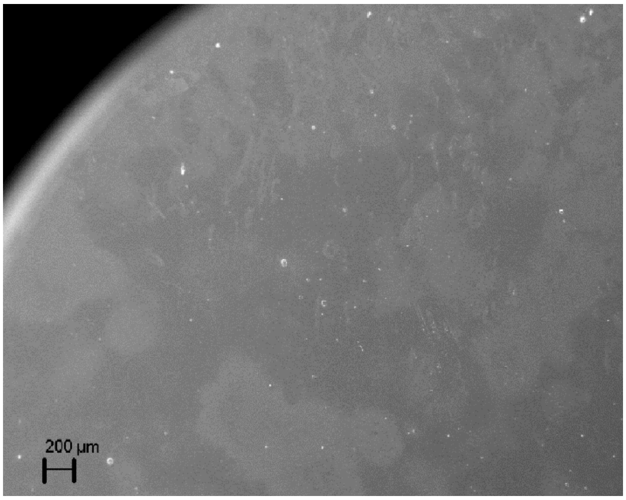

Figure 3: SEM image of a Nb-Pb photocathode with 2 µm Pb film with spherical extrusions.

resulted tight enough to prevent them from reaching the films. It resulted in a presence of a few dozen, unevenly distributed, spherical droplets per mm^2, adjacent to the film surface.

Such protruding surface irregularities of high aspect ratio usually enhance field emission and dark current. It has been verified in two ways: 1. by field emission mapping in dc electric field, performed at HZB on the cylindrical cathode insert of Fig. 3 and 2. by a direct measurement of total dark current at NCBJ in pulsed electric field from a planar Nb-Pb sample. The field emission mapping has been performed in an electrostatic field of average intensity increased from 10 to 25 MV/m by using a field emission scanning microscope (FESM; details of the mapping procedure are available eg in [20]). The dark current emission onset at 14.5 MV/m was accompanied by activation of distinct, local emitters which could be identified with the abovementioned surface protrusions. It is relevant for the application in XFEL-type sc guns operating with the field amplitude of 40-60 MV/m. Fig. 4 shows the emission map at 17.5 MV/m. Field enhancement factors β for selected individual emission centres were found by fitting the measured emission current vs field relationship by Bowler-Nordheim equation. β value in the case of the strongest emitter reached ≈140. Such high β is usually associated with a parasitic increase in both, dark current and thermal emittance. The dark current from certain single emission centres exceeded 10 nA. While new emission centres were activated with rising field some of the active ones disappeared at a sharp drop of local current. This extrusion removal can be treated as one of the mechanisms of cathode surface conditioning in electric field.

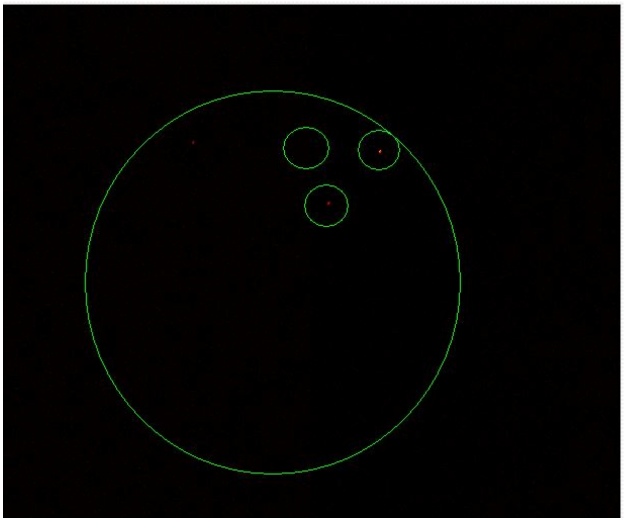

Figure 4: Electron field emission map at 17.5 MV/m on the surface of Pb film shown in Fig. 3 (courtesy of HZB). The big circle corresponds to the cathode perimeter.

Dark current measurement on the planar Nb-Pb cathode sample was performed in a setup with the sample separated by a 100 µm gap from a flat copper anode. Pulsed electric field of 60 MV/m amplitude, 40 µs pulse duration and 0.005 duty factor has been applied between them by using a 12 KV voltage pulser. Voltage drop connected with dark current flow through a 683 kΩ resistor has been measured with an oscilloscope. Dark current dropped by a factor of 4 due to 30 min. long conditioning in the pulsed electric field. surface shown in Fig. 1, proved much lower (≈45 nA) in spite of much higher density of surface defects. It can be attributed to lower aspect ratio of those defects (the ratio of the extrusions height to diameter is below 0.2 for this surface). Our further efforts were dedicated to ex-situ smoothing of thick, non-filtered Pb layers.

SMOOTHING OF PB FILMS IN PULSED PLASMA ION BEAM

Re-melting and recrystallization of Pb films with rough surfaces has been proposed for their flattening. From the available methods of surface engineering we selected pulsed plasma ion beam bombardment of a surface in the rod plasma injector (RPI) IBIS at NCBJ. Its schematic view is shown in Fig. 5 and its detailed description can be found in [21]. The device is composed of two sets of electrically isolated, coaxially arranged tungsten rods. After admitting 1 cm^3 STP of gaseous argon and applying the voltage of 28 kV between them, 1 µs long pulse of Ar ions is created and accelerated to energy ranging from a few kV to 80 kV. The ion pulse reaches the target with a Nb-Pb sample. The value of the pulse energy fluence is monitored by using a system of thermocouples installed in the target support.

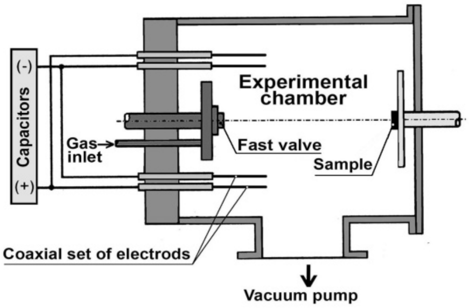

Figure 5: Schematic of the rod plasma injector IBIS.

The first attempts to smooth 1-3 µm thick Pb films with 1-2 J/cm^2 ionic pulses resulted in their perforation. It happened due to direct contact between molten lead and niobium surface. Poor wettability by Pb of different metallic substrates can be attributed to the increase of the surface energy connected with the formation of oxide film on the surface of molten lead [22, 23]. A 2D model for 1 µs heat pulse flow across Pb layer was proposed as reported in [19]. It allowed computation of Pb melting depth and the time of subsequent solidification of lead. It was used to match the primary film thickness with ion pulse fluence so as to melt only the outermost part of the lead film and avoid the direct contact of liquid Pb with niobium substrate. The computation results combined with melting tests in RPI IBIS led to an effective smoothing procedure for 18-20 µm thick Pb films of surface morphology shown in Fig. 1. After treating the film depicted in this figure with five argon ion pulses with 1.5 J/cm^2 in fluence, a smooth lead layer surface has been reached as shown in Fig. 6.

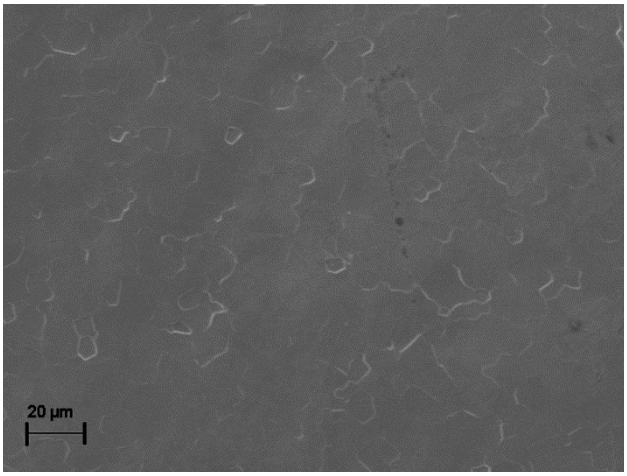

Figure 6: SEM image of the lead film shown in Fig. 1 after treating with five 1.5 J/cm^2 ion pulses in the rod plasma injector.

QE OF OPTIMIZED PHOTOCATHODE AND NB-PB CATHODE IMPACT ON SRF E$^-$ GUN PERFORMANCE

The most important functional features of the discussed photocathodes include their quantum efficiency and the quality factor Q_o of the complete electron gun resonator with the installed cathode.

Nb-Pb sample cathode with 18 µm lead film, prepared according to the optimized ex-situ smoothing procedure described in the previous section, has been tested at BNL for its quantum efficiency in a device based on deuterium lamp with a monochromator. Preliminary, in-situ laser cleaning of the lead emitter surface has been performed in accordance with the rules elaborated in [24]. They provide surface treatment with a pulsed UV radiation 248 nm in wavelength, at a pulse length of 5 ns and repetition rate of up to 20 Hz. At a pulse fluence below 0.26 mJ/mm^2 no changes of Pb surface are observed. The discussed Nb-Pb hybrid photocathode underwent treatment with 10^4 laser pulses of the above characteristics with a pulse fluence of 0.06 mJ/mm^2. The measured quantum efficiency after cleaning was 2.2 x 10^{-3} at a wavelength of 200 nm [19]. It did not differ much from theoretical predictions based on the Spitzer three step model (3.4 x 10^{-3}).

Installation of a Nb-Pb photocathode plug into a gun resonator requires precise vacuum sealing and connection with the liquid helium cooling system. After the cathode installation the resonator undergoes high pressure rinsing in ultra-pure water at a pressure of 100 bar. Due to a limited lead layer robustness against water jet the film had to be protected with a stiff shield for the time of rinsing. Quality factor of a 1.6 cell SRF gun resonator with the optimized Nb-Pb cathode plug, measured in a vertical test performed at DESY, reached $Q_o = 5 \times 10^9$ at a cathode peak electric field of 32 MV/m. This result is inferior (though still acceptable) to the results of a baseline test performed with the same resonator equipped with an uncoated Nb plug ($Q_o = 5 \times 10^9$ at a cathode peak field 52 MV/m). This degradation of the gun structure behavior with Nb-Pb insert may be attributed to the casually enhanced field intensity at the cathode position due to irregularities in the shape of an indium gasket used for vacuum sealing.

SUMMARY AND OUTLOOK

In the recent years various ways of preparation of sc, hybrid Nb-Pb photocathodes have been proposed at NCBJ. A series of tests carried out on photocathode samples, including the measurement of quantum efficiency and dark current along with surface morphology analysis led to the choice of a two-step procedure for preparing the Pb surface film which, for the time being, assured the best operation parameters of the TESLA – type superconducting electron gun. The procedure is composed of the following steps: 1. lead film deposition in a non-filtered cathodic arc on a niobium insert and 2. ex-situ lead layer smoothing in a rod plasma injector by repeated melting with five - six,1 µs long pulses of argon plasma ions with fluence 1.5 J/cm^2 each. This value of energy density ensures effective flattening of the Pb layer without the risk of its perforation. It has been predicted on the basis of a 2D model of heat flow through the film, resulting from the absorption of a single ion beam pulse. QE measurement on the optimized photocathode sample and vertical RF tests on 1.6 cell SRF e$^-$ injector with a hybrid Nb-Pb photocathode gave acceptable results. Due to the widespread desire to minimize the emittance of photoelectron beams accelerated in FEL-s linacs, our team at NCBJ is working on further improving the smoothness of Pb films.

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