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METAL PHOTOCATHODES PREPARATION FOR COMPACT LINEAR ACCELERATOR AT DARESBURY LABORATORY

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Abstract

The photoinjector of the CLARA FEL test facility Front End at Daresbury Laboratory is based on a S-band 10 Hz photocathode RF-gun operating with a copper photocathode which is driven by the third harmonic of a Ti:Sapphire laser (266 nm). The main aim of this study was to establish a procedure to prepare the Cu surface prior to installation so a Quantum Efficiency (QE) of 10^{-5} or higher can be achieved at laser power density below the ablation threshold of copper. In order to do this Cu test samples with a roughness of 10 nm were prepared using different techniques. The best results have been obtained by ex-situ chemical cleaning. This removed the surface oxide layer whilst at the same time producing a surface buffer layer. This inhibited the regrowth of native oxide for up to a week when exposed to normal ambient atmospheric conditions. With either chemical cleaning or Ar plasma cleaning after heating the sample in-situ to 150 °C for 90 minutes or 250 °C for 40 hours, almost all of the surface oxide was removed. For these surfaces a QE of 4.10^{-5} or better was measured. Oxygen plasma cleaning at 100% and 20% power produced CuO layer with surface carbon contaminant to 3 atomic %, however in-situ thermal cycling resulted in at best a QE of $3\cdot10^{-6}$.

INTRODUCTION

The performance of a FEL is strongly related to the brightness of the electron beam generated by the photocathode. The performance of the cathode itself will be strongly dependent on its preparation prior to its installation in the gun. The CLARA accelerator [1], currently under development at the STFC Daresbury Laboratory, will eventually drive a FEL test facility. At the commissioning stage, the CLARA Front End photoinjector is based on a 2.5 cell S-band photocathode RF-gun, operating with a copper photocathode and driven by the third harmonic of a Ti:Sapphire laser (266 nm) [2]. It is installed in a dedicated thermally stabilised room. Light pulses with energies of up to 85 μJ are focused to a spot size of 1 mm with a pulse length which currently can be varied between 2 to 20 ps FWHM and a repetition rate of 10 Hz (eventually 400 Hz). The photocathode for the RF-gun is a polycrystalline, oxygen-free, copper disc machined to 3 nm roughness. It forms an integrated part of the 2.5-cell gun cavity and is the back wall of the first half-cell.

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Once the cathode is installed, in-situ access to it is only available through the 15 mm electron exit aperture. This makes it virtually impossible to process the cathode's surface in-situ without contaminating the gun cavity. To achieve an acceptable QE it is therefore necessary to establish a cleaning procedure which will produce a surface with the lowest achievable work function, whilst at the same time, minimising any changes to the surface topography, prior to installing the cathode into the gun. We have previously reported on a similar study in which we identified that Oxygen plasma cleaning was the preferred procedure within the chosen parameters at the time. However during actual gun operation it was noted that the measured QE of $\sim10^{-6}$ was consistently lower than expected. In order to achieve the desired electron bunch charge we had to increase the laser power density to the point where it severely damaged the cathode surface. To achieve a low emittance the cathode is diamond turned to 3 nm average roughness and roughening the surface with a laser during its operation has to be avoided. Hence a second cathode preparation study was deemed to be necessary to reach a desirable practical solution. Similar studies have been carried out by D.T. Palmer [3,4] and P. Davis *et al.* [5] where they reported QE ranging from 2.5×10^{-4} to 9×10^{-5} for polycrystalline copper.

EXPERIMENTAL PROCEDURES

Polycrystalline oxygen free copper test samples with a surface roughness of 10 nm were degreased with Acetone, IPA and de-ionised water. Four samples were then prepared. One sample was ultrasonically washed in 100% BPS172 solution for 10 minute and then followed by a rinse with Ethanol and finally rinsed with deionised water. BPS-172 is a proprietary copper oxide remover that is commonly used in the semiconductor industry for metal contacts and interconnects [6].The other three samples were plasma cleaned using a Henniker plasma HPT-200 model for 20 minute in 100% Argon or Oxygen atmosphere with the later cleaned at 100% and 20% power settings. All of these steps were carried out prior to introducing samples into vacuum. The surface topography was determined ex-situ by Atomic Force Microscopy AFM.

The QE was calculated by measuring the photocurrent extracted from the sample, *I*, and the power of the incident light, *P* using the Eq. (1):

$$QE = \frac{hc}{\lambda e} \cdot \frac{I}{P} \quad (1)$$

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Where I is the photocurrent (A), h is Planck's constant, c is the speed of light, λ is the wavelength of incident light (m), e is the Electron charge and P is the power of the incident light (W).

An extraction plate placed above the cathode was biased at up to -99 V with a battery box. The cathode was kept at ground potential and the photocurrent was measured with a Keithley type 480 picoammeter.

To achieve photoemission, a compact UV laser (Crylas FQSS 266-Q4) was used. The laser beam was pulsed using a Q-switch and delivers a 1 kHz repetition rate of approximately 1 ns pulses with an individual pulse energy of more than 12 mJ, focused to a spot size of ~ 3 mm FWHM. The average UV power was measured to be around 6.2 mW using a calibrated photodiode.

In order to determine the chemical composition of the copper surface after surface preparation (chemical cleaning or plasma cleaning), samples were transferred into an ultrahigh vacuum chamber and analysed using X-Ray Photoelectron Spectroscopy (XPS). The base pressure of the surface analysis chamber of the XPS system was in the low 10^{-10} mbar during analysis. Photoelectrons were excited by using non mono-chromated Al Kα X-ray irradiation (1486.6 eV), and the photoelectron spectra were taken using a VG Alpha 110 hemispherical analyser at electron emission angle of 90˚ with respect to the sample surface with a pass energy of 20 eV (ultimate energy resolution of 1 eV) for the detailed region spectra and 50 eV for the wide region scans. These pass energies correspond to Ag $3d_{5/2}$ FWHM of 1 eV and 1.2 eV, respectively.

RESULTS AND DISCUSSION

Figure 1 represents the XPS survey spectra of copper samples under different processes. The pure copper surface is represented with two groups of peaks Cu $2p_{3/2}$ at 933 eV and Cu $2p_{1/2}$ at 953 eV due to spin–orbit coupling. The spectra for the samples treated with Oxygen plasma for both powers (100% and 20%) exhibit creation of stoichiometric CuO at the surface with a very small amount of residual Carbon. The Cu $2p_{3/2}$ XPS spectra revealed that the ''as received'' copper surface (Fig. 1d) consists mainly of cupric (Cu^{2+}) oxides as indicated by the shake-up satellite structure centred at a binding energy of 943 eV. The presence of this satellite structure has been attributed to charge transfer transitions from the ligands (O^{2-} ions for CuO) into the unfilled (d^9) valence level of the Cu^{2+} ion [7]. For cuprous oxides (Cu^+), which have a filled (d^{10}) ground state configuration, such transition cannot occur and no satellites are seen (Fig 1a and 1b). For the samples treated with Ar plasma, the cleaning agent (BPS172) and all heat treated samples, the satellite peak is not present, however it is not clear whether the peak at 932.67 eV is due to metallic copper or Cu_2O because the Cu $2p_{3/2}$ binding energies and peak widths of these two species are very similar. The broader peak shape of the cupric species as compared to the Cu/Cu_2O peak is due to coupling between unpaired electrons (multiplet splitting) in the paramagnetic cupric ions [7, 8]. Oxygen O1s and carbon C1s are represented by peaks at 532 eV and 284 eV respectively and their atomic percentages are presented in Table1.

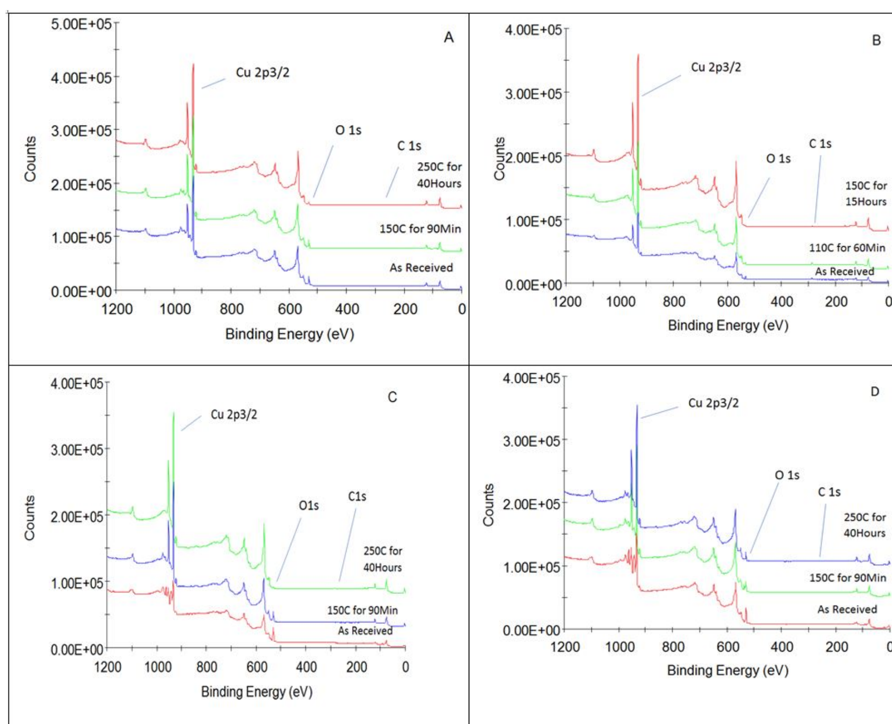

Figure 1: XPS survey spectra of as received and in-situ heat cycle for different cleaning procedures a) Ar plasma cleaned, b) BPS172 chemical cleaned, c) O Plasma Cleaned at 20% power and d) O_2 plasma cleaned at 100% power.

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Table 1: Atomic fraction and peak position of Cu, O, and C as determined by XPS analysis and QE analysis of the copper samples before and after the experimental cleaning procedures.

	O plasma 100% power			O plasma 20% power			Ar plasma 100% power			BPS 172 treatment		
	As treated	**150°C** 90 min	**250°C** 40 hrs	As treated	**150°C** 90 min	**250°C** 40 hrs	As treated	**150°C** 90 min	**250°C** 40 hrs	As treated	**110°C** 60 min	**150°C** 15 hrs
Cu $2p_{3/2}$	49.0 % 933 eV	48.0 % 932 eV	66.3 % 932 eV	33.9 % 933 eV	47.9 % 932 eV	72.1 % 932 eV	47.7 % 932 eV	74.7 % 932 eV	80.0 % 932 eV	40.0 % 932 eV	69.0 % 932 eV	80.0 % 932 eV
O 1s	47.6 % 529 eV	33.7 % 530 eV	33.7 % 531 eV	55.8 % 530 eV	40.0 % 530 eV	15.5 % 531 eV	47.1 % 530 eV	22.1 % 532 eV	17.2 % 532 eV	32.0 % 530 eV	15.0 % 532 eV	10.0 % 532 eV
C 1s	3.4 % 284 eV	0	0	10.3 % 284 eV	12.1 % 284 eV	12.9 % 284 eV	5.3 % 284 eV	3.2 % 284 eV	2.7 % 284 eV	28.0 % 284 eV	16.0 % 284 eV	10.0 % 284 eV
QE	$>10^{-7}$	$>10^{-7}$	$>10^{-7}$	$>10^{-7}$	$>10^{-7}$	3.0×10^{-6}	1.5×10^{-7}	3.0×10^{-7}	3.0×10^{-5}	1.5×10^{-5}	4.5×10^{-5}	4.5×10^{-5}

It can be seen that carbon is only present in significant amounts when the sample has gone through 20% power O_2 plasma cleaning or the cleaning agent solution. In the latter case the exact nature of the carbon at the surface cannot be determined with the current energy resolution of the experimental set up. It is thought that this is a carbonate or hydrocarbonate buffer layer that protects the surface from becoming oxidised when it is exposed to ambient atmosphere. It further proved to be very resistive to both post O_2 and Ar plasma cleaning. Nevertheless, from the QE results reported in Table 1, it was seen that such a buffer layer does not hinder the achievement of the highest QE in this set of experimental results.

In Plasma ions, radicals and UV radiation are formed at the same time and can act all together. The O_2 plasma cleaning enhanced the oxidation process and created a fully oxidized surface. It is believed that the majority of hydrocarbon cleaning is performed through chemical reaction of oxygen free radicals, created in the plasma, rather than direct ion impact sputtering of the surface contaminant layer. The longer wave length UV radiation (>200 nm) that is not absorbed by the oxygen atoms will be strongly absorbed by the organic chemical bonds present on the surface of the substrate. Upon absorption this causes the formation of excited states or organic radicals. These reactive organic species and oxygen free radicals result in the formation of volatile species such as CO_2, H_2O, N_2 and short chain organic compounds. These volatile compounds can easily desorb from the surface leaving it clean. However, in Ar plasma, the cleaning process is mostly taking place by ion sputtering.

In-situ heating of the BPS172 and Ar plasma cleaned samples to 110 °C for 60 min and 150 °C for 90 min respectively led to significant reduction on the amount of Oxygen present on the surface, presumably by dissolution into the bulk of the sample. In the case of BPS172 the oxygen reduction was accompanied by an increase in QE to desired levels (4.5×10^{-5}). In the case of the Ar plasma heating in-situ to 250 °C for 40 hours reduces the oxygen even further with concurrent increase in the similar measured QE (3.0×10^{-5}). For the 20% power O_2 plasma cleaned sample 40 hours at 250 °C were needed to achieve a decade lower QE and for the 100% power O_2 plasma cleaned sample not even this was sufficient to generate any significant QE. Annealing to 250 °C for 40 hours was chosen since this is the highest temperature that can be achieved when baking the RF-gun after installation on the accelerator. In addition 40 hours represents a typical baking time.

These results are to some extent contradictory to previous work on O_2 plasma cleaned Cu where 250 °C annealing was seen to reduce the O level at the surface and produce reasonable QE [9]. Previous data was collected for samples prepared using a different apparatus and it would appear that different plasma cleaners do not necessarily produce the same results. At the present time Ar plasma treatment is being used to produce operational photocathodes for the CLARA front-end injector since this technique has been demonstrated to produce sufficient QE. However, the BPS172 treatment appears to be a promising alternative and further investigation is planned.

CONCLUSION

The highest QE was achieved using BPS172 and short, low temperature (150 °C) annealing solution where the oxide is removed and a protective hydrocarbon or carbonate layer is formed. Ar plasma cleaning achieved similar QE results, however it requires a longer annealing time at a higher temperature (250 °C). Oxygen plasma cleaning produces a stable CuO which requires a high temperature (400-500 °C) annealing to achieve high QE. Ar plasma treatment is currently being used to prepare photocathodes for the CLARA front-end.

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