Low Workfunction Surface Coatings for Dispenser Photocathodes in Radio Frequency Photoinjectors

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Photocathodes are a critical component in photoinjectors for free-electron lasers (FELs) and other accelerator applications requiring a high-current, low-emittance electron beam. An ideal photocathode would have high efficiency in the visible range, a long operational lifetime in a typical accelerator vacuum environment, and prompt electron emission. Efficiency is typically improved by adding a photosensitive, cesium-based compound to the cathode surface. Because this layer is chemically active, however, it is vulnerable to evaporation and contamination, causing the cathode to degrade with use. Reapplication of this surface layer requires operational downtime and motivates the need for an in situ rejuvenation technique that would prevent the cathode from being a significant failure mode of an FEL system. This work determines the effect of surface cesium coverage on the efficiency of a metallic cathode and compares theory and experiment as precursory work toward a dispenser photocathode. A prototype multialkali dispenser photocathode is proposed, along with a measurement technique used to monitor its surface conditions during operation.

Nomenclature

i	electron beam current, A
P_λ	laser power, W
QE	quantum efficiency, %
x_{exp}	thickness, Å
x_{max}	coating thickness at peak QE, Å
α	deposition rate scale factor, Å^{-1}

θ coverage, %
λ laser wavelength, μm
ϕ workfunction, eV

1. Introduction

The free-electron laser (FEL) is being pursued as an all-electric weapon system offering variable lethality, wavelength selectivity, and megawatt power capability.[7] The required high-current electron beam originates at a photoinjector, where drive laser pulses impinge upon the photocathode at intervals corresponding to the accelerating phase of the radio frequency (RF) field.[8] The photocathode then emits an electron beam whose pulse shape ideally corresponds to that of the drive laser. This arrangement offers the advantage of both high beam quality and high current. Photocathodes are problematic, however, because they exhibit low efficiency, short lifetime, and a vulnerability to fluctuating vacuum conditions.[6] Additionally, characteristics of the drive laser are generally at odds with those of the photocathode: the highest cathode efficiencies occur at wavelengths that are currently unattainable for a sufficiently stable drive laser.[5]

Quantum efficiency (QE) is an important figure of merit and is defined as the ratio of emitted electrons to incident photons. Many materials have been investigated for use as photocathodes.[6,10] The QE for most cathode materials is quite low and is improved by a surface coating (e.g., cesium) that reduces the workfunction ϕ, the energy required for an electron to overcome the surface barrier to vacuum. This coating is usually on the order of a monolayer of atoms in thickness and is easily damaged by desorption, contamination, and/or ion back-bombardment. The lifetime of the photocathode is therefore determined by the longevity of this surface layer of photosensitive material. A short cathode life would severely limit the reliability and duty cycle of an FEL weapon system. The ideal cathode would have a self-repairing surface with high QE in the visible to accommodate the present limitations in drive laser technology. This work specifically addresses the challenges relating to cesiated photocathodes, while other groups focus on drive laser improvement.[1]

The dispenser concept has its origins in the microwave tube industry, in which its application to thermionic emission is crucial to achieving high reliability. In such cathodes, a compound containing alkali metals is buried in a host matrix material, such as tungsten. The high temperature at which the cathode operates then causes this material to diffuse from within the bulk to the surface at some controlled rate, at which it reduces the workfunction and leads to enhanced electron emission. Thermionic cathodes yield low current densities, however, which lead to complexities in FEL design (such as bunching cavities). Another concern is ion back-bombardment, which occurs during the decelerating phase of the RF cycle. For these reasons, photoemission (rather than thermionic) is considered more applicable for FEL systems.

To adapt the dispenser concept to a photocathode designed for high QE, a subsurface reservoir of cesium chromate in powder form is situated beneath a sintered tungsten disk 1 mm thick. During an activation process, elemental cesium is produced, which then diffuses to the cathode surface at a rate determined by the sintered porosity and operating temperature. This design, in principle, can accommodate almost any cathode material, including semiconductors, which require the presence of cesium to achieve a sufficiently low workfunction. It is important to note that the activation temperature ($\sim$500°C) is much higher than the planned operating temperature.

Designing and fabricating a functional photocathode could involve an extraordinarily large amount of empirical work, because each candidate compound and composition could yield a completely different cathode. In the case of ternary compounds, typical of semiconductor cathodes with higher QE and longer lifetime, the number of unique combinations becomes quite large. For this reason, a comprehensive amount of experimental data for a simple cathode (cesiated tungsten) was obtained in this work and a theory was devised to account for the photoemission observed.[3] By using material properties of different component materials, the theoretical behavior of other photocathodes can be explored without having to build each one individually. Only those that appear promising from the theoretical standpoint are pursued experimentally. For example, the behavior of cesiated silver was predicted and then shown to agree well with experiment. The most important step in predicting photocathode behavior is to determine workfunction variation with surface coating and how this, in turn, affects the photocurrent and quantum efficiency. Because photoemission from coated metals is largely a surface effect, the surface conditions of the cathode are paramount. Therefore, determining surface conditions and the effect of coatings on QE are the main objectives of the research presented here, with the overall goal to design and build a robust, long-lived, efficient photocathode.

2. Photocathodes

2.1. Basics

Photoemission can be thought of as occurring in three steps, each of which illustrates the challenge in designing an optimal photocathode.[12] These are photon absorption, electron migration to the surface, and barrier interaction. Because of the characteristically high optical reflectivity of metals, photon absorption in such cathodes is inherently limited. Semiconductors are less reflective than metals, and consequently more incident photons are absorbed (if their energy exceeds the bandgap).[11] This contributes to the low QE of metallic-based photocathodes and the higher QE of semiconductor-based photocathodes.

Step 2 involves the process by which a photoelectron travels to the surface. An electron that has absorbed a photon is considered hot because its energy is higher than that of other electrons in thermal equilibrium. Interaction between this "photoelectron" and other electrons (or the crystal lattice) will reduce the chances of its arriving at the surface with sufficient energy to cross the barrier. Because of the abundance of free electrons in metals, photoelectrons created deep within the bulk will undergo many collisions with other electrons and rapidly thermalize. Therefore, only electrons excited within a few atomic layers of the surface will arrive with sufficient energy to escape and contribute to photoemission. Because virtually no transit time is required for electrons to cross the barrier in metals, such photocathodes are considered prompt emitters. Prompt emission (less than several picoseconds) is generally considered a requirement for FEL photoinjectors.

In semiconductors, the dominant scattering mechanism is electron–phonon interaction, involving much less energy than electron–electron interaction. Consequently, photoelectrons in semiconductors can travel much greater distances (compared with electrons in a metal) before reaching thermal equilibrium. Together with a longer photon penetration depth in semiconductors, this means that more photoelectrons will cross the barrier potential, yielding higher QE. The transit time for electrons excited deep within the bulk, however, introduces a delay in emission with respect to the incident laser pulse.

The third step is barrier interaction. For metals, the surface barrier is simply the workfunction, typically between 4 and 5 eV. This photon energy corresponds to UV light, which

is not practical for FELs because of drive laser limitations.[5] This illustrates the typical tradeoffs with photocathodes: metallic types are prompt emitters, robust (long lived), and simple, but have low QE and require unpractical drive lasers. Semiconductor types have high QE but are not prompt emitters and are delicate and more complicated to build. This research attempts to find a compromise by extending the lifetime of moderately high-QE photocathodes using the cesium dispenser concept.

2.2. Megawatt-class FEL requirements

From the standpoint of FEL design, the drive laser and the photocathode are not independent. The reliability of the drive laser directly impacts the reliability of the FEL system as a whole. This is why a cathode with high QE in the visible is desired: UV lasers (frequency quadrupled from a $\sim$1-μm fundamental via nonlinear crystals) are not considered reliable for high-duty-factor operation. For a megawatt-class FEL, approximately 1 A of average beam current is required given a beam energy of 100 MeV and an extraction efficiency of 1% (1 A $\times$ 100 MeV = 100-MW beam). Extraction efficiency is the ratio of outcoupled FEL power to electron beam power. Average electron beam current depends on drive laser wavelength, drive laser power, and quantum efficiency:

$$i[A] = \frac{\lambda[\mu m]}{124} \cdot P_{laser}[W] \cdot QE[\%].$$

This means that with green light (532 nm) and a drive laser of 50–100 W, achieving an electron beam of 1 A requires a QE of 2–4%. Photocathodes with this efficiency currently exist but have impractically short lifetimes unless stringent UHV conditions are maintained. The dispenser concept could extend the effective lifetime of such cathodes to provide both the beam current and system reliability required of an FEL photoinjector.

3. Experimental Setup

The experimental facility at the University of Maryland consists of a UHV chamber housing the alkali-metal evaporation sources, an anode/cathode assembly, a deposition monitor, and a cathode heater. The chamber can achieve pressures of about 10^{-10} torr and is equipped with a fine metering valve to intentionally leak trace gases to either assist in surface processing or simulate an elevated background pressure. Quantum efficiency is monitored by shining one of four temperature-stabilized diode lasers onto the cathode in the presence of an electric field while measuring the resulting photocurrent as a function of evaporated thickness or coverage. The lasers emit at the following wavelengths: 850 nm (IR), 655 nm (red), 532 nm (green), and 405 nm (blue), allowing QE measurements at various photon energies. Because photocurrent in this experiment could be as small as tens of nanoamperes, the entire QE circuit was double shielded to lower background noise. The photocurrent signal and pressure, temperature, wavelength, evaporated thickness, background gas composition, and anode–cathode voltage are measured and recorded using Labview-integrated instrumentation.

Figure 1 shows a schematic of the four-way cross and the evaporation system used to fabricate cesium on tungsten (Cs–W) and cesium on silver (Cs–Ag) cathodes. For dispenser cathode testing, the cathode assembly is slightly modified and the evaporation sources are expanded to include elements beyond just cesium. Sources of potassium and antimony will allow the fabrication of more complex and higher-efficiency dispenser photocathodes.
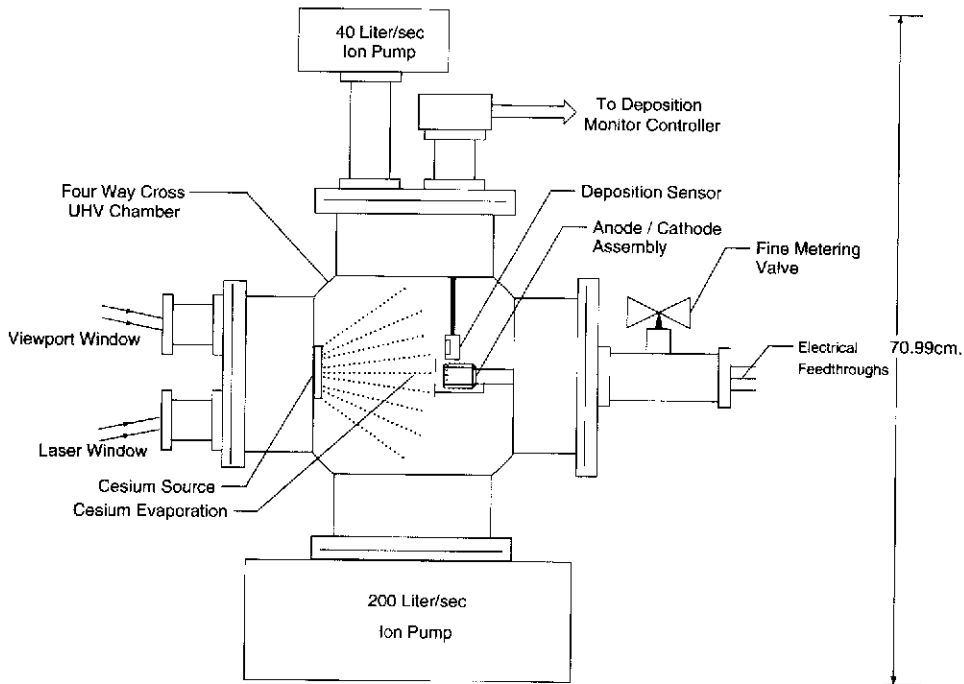
Fig. 1. University of Maryland UHV fabrication system.

To reasonably compare prediction with experiment, surface conditions of the cathode must be as close as possible to those assumed in the theory. Ideally, a cathode such as Cs–W would be built completely in vacuum by evaporating tungsten (or silver) onto a substrate and then depositing cesium on top. In this manner, one can be assured that cesium arrives at a clean metallic surface, free from contamination or oxide layers. This is laborious from an experimental standpoint, so a high-purity metal disk (of either tungsten or silver) was exposed to plasma discharge and high-temperature anneal in order to obtain a reasonably clean surface. Cesium was then deposited on the surface at a constant rate, as shown in Fig. 2, and measured using a crystal balance (Fig. 2, inset).

4. Results

4.1. Experimental

The most striking feature of the results to follow is their close agreement with theoretical prediction. While details of the theory are described elsewhere,[3] it is important to recognize that nearly all factors affecting QE were taken into account. These include laser properties (wavelength, intensity, heating of the cathode), photocathode material (specific heat, thermal conductivity), surface coatings (workfunction variation), and the local environment (electric field, temperature).

Data were obtained by measuring QE as cesium was slowly added to the surface over a period of about 10 min. The deposition monitor reports the thickness of cesium coatings, but it is more logical to express coverage in percent monolayer, since this is independent
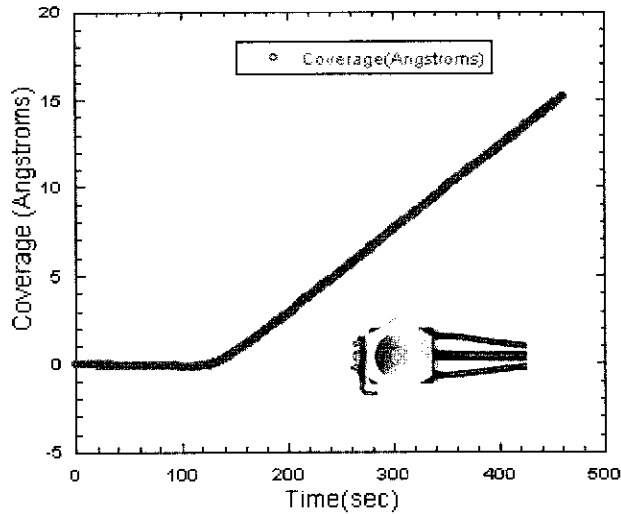
Fig. 2. Cesium deposition as measured by crystal balance (inset).

of atomic species. Percent monolayer is roughly determined by dividing the coating thickness by the covalent diameter (for cesium, 5.5 Å). The Gyftopolous–Levine model for photoemission[2] is used to align the experimentally observed peak in QE with the coverage at which this is expected to occur. By taking into account the deposition rate (which tends to scale the thickness reading by α), the following relationship between thickness θ and coverage x is obtained[4]:

$$\theta(x) = \alpha(x_{exp} - x_{max}) + 63.537\%.$$

The experimental data, together with theoretical prediction, for the case of cesiated tungsten are shown in Fig. 3. Notice that for blue and green (405 and 532 nm, respectively), the theory and experiment agree exceedingly well. Matters are more complicated for red light (655 nm), because the photon energy is approaching the workfunction. Near the barrier height, the photoemission process is very sensitive to any changes on the surface, and it is quite possible that the details of the actual surface do not match those being modeled. Nevertheless, multiplication (of the theoretical prediction for red light) by a factor of 3 appears to bring it back into agreement with experiment. Further investigation is required to determine the need for this factor. Aside from this detail, no other adjustments were made to the theoretical predictions to obtain the agreement shown. The error (for all measurements in this paper) in QE is less than 0.003%, and the error in coverage as read from the monitor is about 0.2%. The peak QE that occurs at all wavelengths corresponds to a fraction of monolayer coverage and is thus a relative indicator of the amount of cesium remaining on the surface. This measurement will serve as a crucial diagnostic in the dispenser cathode phase because it allows estimation of remaining cesium given changes in quantum efficiency.

The comparison of theory and experiment for cesium on silver is shown in Fig. 4. The qualitative agreement demonstrates that the theory developed for cesiated tungsten can successfully predict approximate behavior for other cathode materials. This is a first step toward predicting the behavior of more complicated photocathodes. Notice in Fig. 4 that two
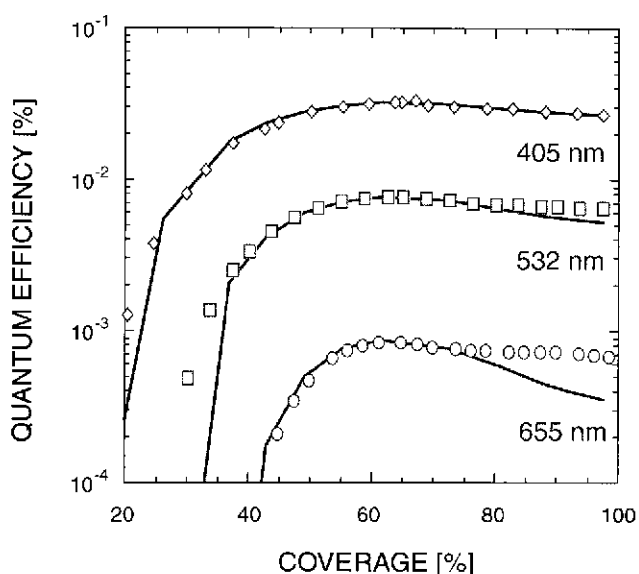
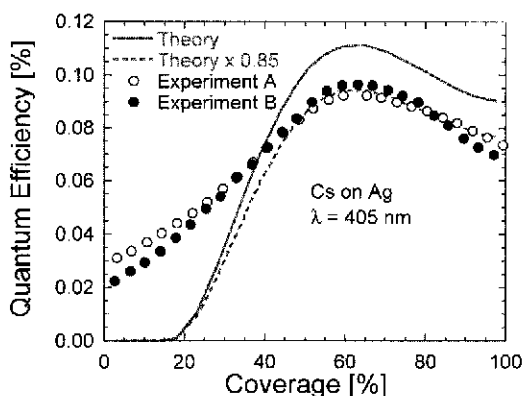**Fig. 3.** QE vs. cesium coverage on tungsten at various wavelengths.

Fig. 4. QE vs. cesium coverage on silver at 405 nm.

sets of experimental data are plotted (discrete points). These represent two experimental data sets obtained on different days, using different cesium sources and separate heat treatments; the fact that they nearly coincide imparts confidence in the repeatability of experimental techniques used. The theoretical QE for Cs–Ag is consistently 15% higher than that observed experimentally. This can be attributed to the difficulty in cleaning the surface of silver. Unlike tungsten, silver cannot be annealed at high temperatures because of its lower melting point, and the degree to which the surface can be cleaned is limited. The discrepancy between predicted and actual QE at low coverage is due to the difficulty of removing cesium completely from silver. (Residual cesium from previous experiments causes photoemission at seemingly low coverage.)

Figure 5 shows QE as a function of time for a cesiated-silver photocathode: over a period of about 1 week, the cathode's QE drops to $1/e$ of its original value. This can be considered

Cs-Ag Lifetime: $T_{1/e}$ =166 hours
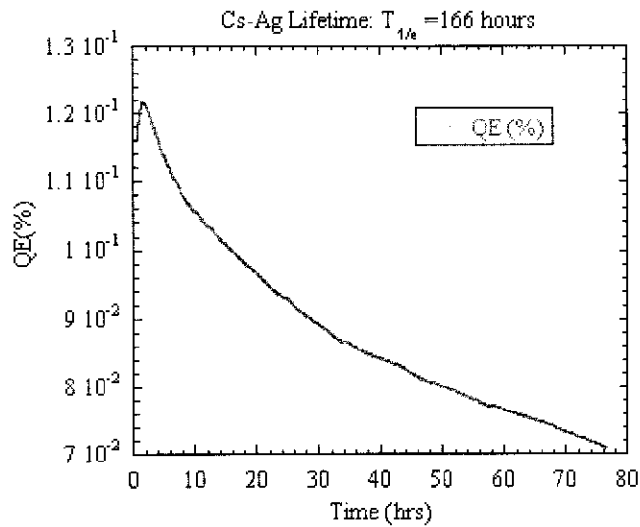
Fig. 5. QE vs. time for cesium on silver photocathode.

its useful lifetime and illustrates the need for a rejuvenation method to restore high QE, namely the controlled-porosity dispenser photocathode.

4.2. Dispenser photocathode

Figure 6 shows a simplified schematic of a dispenser photocathode: a stainless-steel canister is filled with cesium chromate powder and nickel brazed to a sintered tungsten disk on the top. A prototype was recently fabricated using sintered tungsten and is shown to
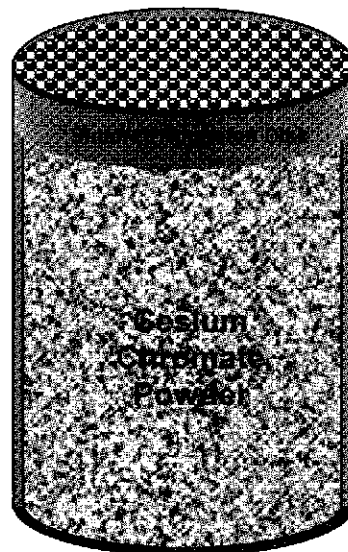
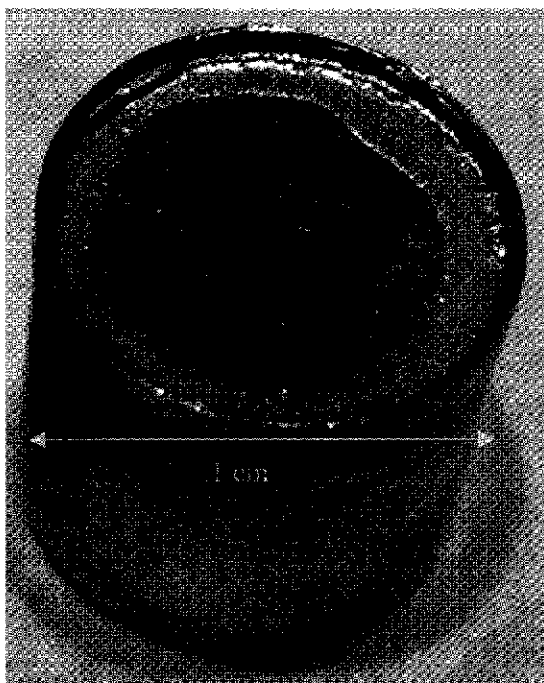**Fig. 6.** Schematic of dispenser photocathode.

Fig. 7. Prototype dispenser photocathode.

scale in Fig. 7.[†] During initial activation heating, cesium chromate is reduced to elemental cesium, which should diffuse to the surface in a controlled manner through the microscopic pores in the tungsten disk.

Figure 8 is a scanning electron micrograph (SEM) image of the dispenser photocathode surface, showing the pores through which cesium will diffuse to cover the surface. Grains, grain boundaries, and pores are discernible, with a closeup of a 0.5-μm-diameter pore shown in the inset. One feature immediately obvious is that spacing between pores is not uniform. Using wider fields of view, the statistics of pore separation were studied and found to follow a log-normal distribution with a mean pore–pore separation of 2.47 μm. Electron emission on the surface depends on workfunction, which, in turn, depends on cesium coverage. Uniform coverage, therefore, is needed to minimize the emittance of the electron beam. Because of irregular pore spacing, coverage will inevitably be nonuniform, and studies are in progress to quantify the surface diffusion of cesium. It appears likely that low workfunction "patches" near the pores will contribute most to the emitted electron beam, surrounded by areas of higher workfunction and lower emission. The pore–pore separation provides the average distance cesium atoms would have to travel along the surface in order to completely cover a given grain.

The average grain appears to have a diameter of approximately 5 μm, found by calculating the area of several representative grains. It has been posited that cesium will arrive at the surface not only via pores, but perhaps also via grain boundaries. For this reason,

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[†]Fabrication performed by B. Vancil, EBeam, Inc., 2005.
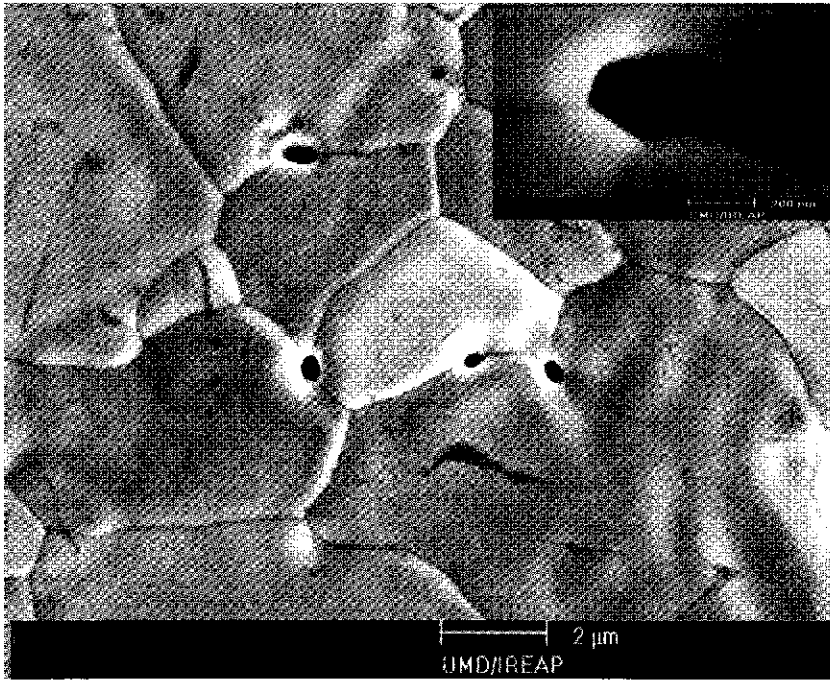
Fig. 8. SEM of sintered tungsten cathode surface (pores and grains).

the three-dimensional structure of a grain was studied using focused ion beam milling. A 30-keV beam of germanium ions was scanned over a 50×50 μm area to slowly remove tungsten atoms, as shown in Fig. 9, while the resulting secondary (backscattered) electrons provided an image. Because grain orientation determines the backscattered signal intensity and because neighboring grains have different orientations, they can be differentiated from one another during the milling process. To determine a grain's three-dimensional (3D) structure, its two-dimensional contour was recorded at the surface and also at 1-μm depths during the milling process. These contours were then assembled as a mesh image in three dimensions. The resulting approximate shape of a representative grain (shown from a subsurface perspective) is given in Fig. 10. Notice the irregular, jagged sides of the grain: this complexity suggests that grain boundaries will not be a dominant cesium delivery mechanism because of the long path lengths involved. Figure 11 also shows that the typical grain size in the z direction (normal to cathode surface) is about 5–6 μm.

4.3. Surface coverage via quantum efficiency

For the measurements described in Sec. 4.1, cesium was deposited on the cathode surface from a source, and coverage could be measured directly using a deposition monitor. In the case of a dispenser cathode, cesium again reaches the surface, but from within the cathode itself. This means that cesium coverage of a dispenser cathode cannot be determined directly using a deposition monitor. Because QE varies with coverage, however, it can serve as a noninvasive measurement technique for determining how much cesium has been brought from the plenum to the surface. The agreement between QE vs. coverage experimental data
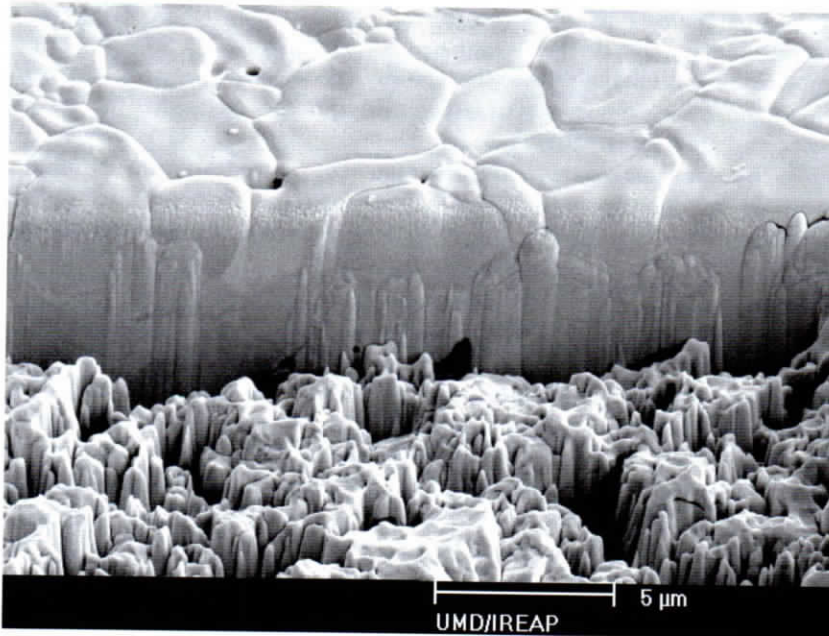
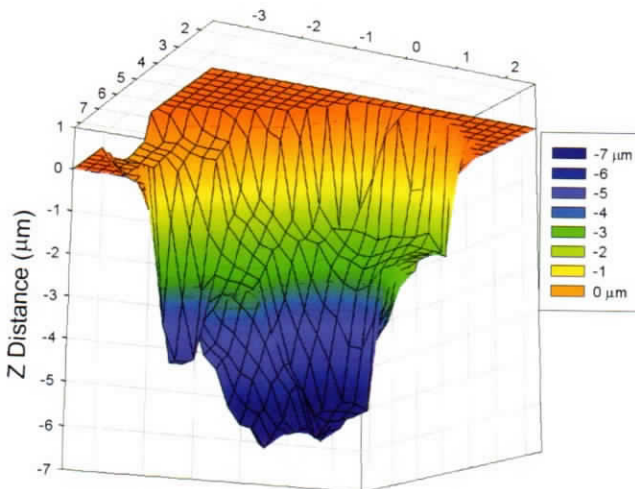**Fig. 9.** Focused ion beam micromachining of photocathode surface.

Fig. 10. 3D shape of sintered tungsten grain.

and theory suggests that QE is a viable measure of surface conditions. Significant effort must still be applied to determine the optimum activation procedure for bringing cesium to the surface at a controlled rate, and this measurement technique will play a crucial role. The peak in QE at submonolayer coverage provides an unambiguous checkpoint: when this peak is reached, about 67% surface coverage of cesium can be assumed.

Sintered tungsten was chosen as a convenient starting point for dispenser cathode research, and it is quite possible that only certain porosities will allow the cesium coverage

required. A given porosity could either restrict diffusion too much or permit it to occur too rapidly (contaminating the RF gun with excess cesium). Operating temperature provides another means of controlling diffusion through sintered tungsten, but a different controlled porosity method, laser drilling, may also be utilized. In this method, pores are drilled at regularly spaced intervals, with desired diameter and separation. While this is more costly than sintered tungsten, the regularity of pores may increase workfunction uniformity, leading to a lower emittance beam. In either the sintered tungsten or the laser drilled case, cesium diffusion across a surface is critical and is being investigated at the Naval Research Laboratory.[9]

5. Summary and Conclusion

FELs show promise as a next-generation light source because they can theoretically generate wavelength-selective radiation at currently unattainable spectra and power levels. A key technology for the development of FELs is the photoinjector, in which the electron beam originates. This research is motivated by the need for robust, long-lifetime photocathodes with high quantum efficiency at visible wavelengths. A novel approach to high QE and long cathode life is the controlled-porosity, multialkali dispenser photocathode: cesium is stored in a reservoir beneath the cathode and diffuses to the surface during a rejuvenation process to replace that which is lost. Cathode porosity and temperature are adjusted such that approximately one monolayer of cesium is maintained at the surface. In preparation for the dispenser approach, the behavior of cesiated metal photocathodes was modeled and experimentally verified. Quantum efficiency as a function of temperature, pressure, cesium coverage, laser intensity, and laser wavelength was measured and found to agree strongly with recent theory. This agreement permits QE to be used as a noninvasive surface diagnostic for dispenser cathodes, useful in optimizing the activation procedure and selecting the operating temperature required to bring cesium to the surface. The cathode surface and 3D grain structure were studied and found to be complex, perhaps leading to patchy electron emission, which could be improved using a laser-drilled cathode substrate. All the cathodes described in this work are admittedly simple and have too low QE to be considered candidates for use in an FEL. The purpose of studying simpler cathodes first is to establish a predictive theory that can be used to select the composition of more complex, higher-QE cathodes and to determine the effects of surface coatings on metals for a dispenser cathode. Recent results (to be published) have shown that the dispenser concept proposed in this work successfully delivers cesium to the surface at a controlled rate and at predictable temperatures. A temperature of 470°C was required to activate the dispenser and create an excess of cesium within the cell. Subsequent heating to temperatures between 160 and 180°C allowed cesium to slowly diffuse to the surface and create a surface layer that yielded a QE of 0.04% at 532 nm. These initial results, together with detailed knowledge of the effects of alkali coatings on cathode substrates, suggest that a long-lived photocathode for FEL photoinjectors could be realized by using the in situ dispenser concept.

6. Acknowledgments

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