Breakdown and field emission conditioning of Cu, Mo and W

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Abstract

The ultra-high-vacuum electrical breakdown characteristics of copper, molybdenum and tungsten have been explored in a setup based on a capacitor discharge. Upon repeated sparking, tungsten and molybdenum showed improvement of the maximum applicable field before breakdown (conditioning) in contrast to copper, which experienced alternate improvement and degrading. After conditioning, tungsten withstood the highest applied field followed by molybdenum and copper. This behaviour was correlated with that of the field enhancement factor $\beta$ extracted from measurements of the field emission current. These results are compared with the tests performed on 30 GHz test accelerating structures for the future Compact Linear Collider.

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1. INTRODUCTION

The requirement for high accelerating gradients in future particle accelerators, including linear colliders such as CLIC (Compact Linear Collider) [1], has recently motivated an extensive study of RF breakdown [2]. The relationship between dc and RF breakdown has emerged as an important question: if the physical processes of dc breakdown [3,4,5] apply to RF breakdown, the relatively inexpensive and easily instrumented dc experiments could be used to predict RF behaviour. This report summarizes initial experiments on vacuum dc breakdown made from this new perspective.

The goal of CLIC is to produce multi-TeV $e^+e^-$ collisions through accelerating gradients of the order of 150 MV/m, which are necessary to limit the machine to a reasonable length but imply surface fields of 400 MV/m. Such a gradient is well beyond that used in existing accelerators and development programs in dedicated test facilities [6,7] are underway. Extensive damages due to RF breakdown [8] and a frequency independent surface electric field limit [9] have been observed in copper accelerator structures. Increasing the achievable gradient by using refractory metals rather than copper has been proposed [10] and accomplished [11]. The RF devices in these experiments are large and complex and require substantial resources to run, hence our interest for simpler dc simulations.

In this paper we present the results of experiments performed with a dc spark-test system [12], which have been carried out on copper, molybdenum and tungsten, the candidates investigated in the 30 GHz experiments described in Ref. [11]. The breakdown field is measured as a function of the number of spark events in order to characterize the ageing behaviour of the electrodes and to compare it to that induced by RF operation. Additionally, the field emission current as a function of the applied dc field is measured, since it is established that breakdown processes are related to the field emission behaviour of the electrodes [4] and energy losses in present accelerator test facilities [8] could be related to RF interaction with the field emission currents.
2. EXPERIMENTAL METHOD

The dc spark-test system used for the experiments is described in detail in another report [12] and a schematic view is presented in Fig. 1. In short, the electrodes are located in ultra high vacuum, at a pressure below $3 \times 10^{-9}$ mbar after bake-out at 150 °C. The anode is a mechanically rounded hemispherical tip with diameter of 2.4 mm placed in front of a flat plate acting as the cathode. UHV-compatible translation devices enable control of the electrode distance at μm accuracy level and allow lateral translation to perform measurement on several sites on a sample. Both the cathode (plane surface) and the anode (hemispherical tip) are made of the same material. In the present investigation we use OFE copper, W (99.95 % purity) and Mo (99.9 % purity). All the electrodes were degreased by the standard CERN chemical cleaning procedure [13] before insertion in UHV.

The electronics for data acquisition, application and switching of the high voltage is completely computer controlled. The high voltage (up to 12 kV) is applied to the electrode gap through a charged capacitor. The rise time of the voltage upon application to the junction is about $4 \times 10^{-7}$ s, with a fast ($2 \times 10^{-7}$ s) decreasing overshoot of about 15 % of the nominal voltage. The total available energy stored in the capacitor is 1.4 J at 10 kV. The capacitor voltage is applied to the electrode gap and the decrease of the capacitor charge from its initial value is monitored after a fixed time (2s). This can occur either through field emission currents or through a breakdown event in the gap. At voltages and fields lower than the threshold leading to breakdown, the behaviour of the charge on the capacitor can be derived analytically [12] based on the field emission current equation. So-called spark-scans [12] are carried out, where the high voltage on the capacitor is increased stepwise and the corresponding charge decrease is monitored for each voltage up to the occurrence of a spark. A true spark, or breakdown, is recognized from its characteristic current signal detected by a Rogowski coil wound around the connection from cathode to ground. The field in the gap is calculated with a plane electrodes approximation and the distance between the electrodes is measured via the calibrated translator holding the tip. With this dc setup the breakdown field $E_{b1}$ can be measured from spark-scans and a sequence of them is used to monitor the ageing behaviour of the electrodes.
Field emission current measurements are performed with the same setup, but with the high voltage applied directly by a power supply, the current being limited by a series MOhm resistor. The field emission current is given by the Fowler-Nordheim equation for field emission [3]:

\[
I = A_e \frac{1.54 \times 10^6 \beta^2 E^2}{\phi} e^{10.41 \phi^{-1/2}} e^{\frac{10^5 \phi^{3/2}}{E}} \equiv \bar{\xi} E^2 e^{-\gamma/E}
\]

where \( I \) is the field emission current in A, \( E \) the electric field in MV/m, \( A_e \) the emission area in \( m^2 \), \( \phi \) the work function in eV, \( \beta \) the dimensionless field enhancement factor and the constants \( \gamma \) and \( \bar{\xi} \) are material and geometry dependent parameters. This equation is valid when temperature corrections [14] and space charge limitations [15] to field emission currents are neglected.

In a logarithmic representation the equation reduces to the usual FN plot:

\[
\ln\left( \frac{I}{E^2} \right) = \ln(\bar{\xi}) - \left( \frac{\gamma}{E} \right)
\]

so that from the slope of the straight line we can extract the \( \gamma \) value and hence \( \beta \) as:

\[
\beta = \frac{6.53 \times 10^3 \phi^2}{\gamma} = \frac{62335}{\gamma}
\]

For simplicity we set \( \phi = 4.5 \text{eV} \), throughout this article, which is within the range of literature values for all the materials investigated here [16]. Initially, work function changes due to adsorbates, oxide, crystalline face, or polycrystallinity are neglected. The local field at the field emission site is then \( E_{\text{loc}} = \beta E \).

3. RESULTS

In the present investigation surface ageing is monitored by measuring the (applied) breakdown field \( E_{b1} \) for many subsequent spark-scans at the same site of the sample. Typical conditioning curves for Cu, Mo and W are presented in Fig. 2. A clear difference is visible between the various materials. W shows an almost monotonic increase of the breakdown
field, which reaches values of 500-600 MV/m, whereas for Cu the value of $E_{b1}$ never exceeds 300 MV/m and rather oscillates below 250 MV/m without clear improvement as a function of the number of sparks. Mo has a similar behaviour as W, with a monotonic increase of the breakdown field, but the conditioning is slower and possibly saturates at lower $E_{b1}$. In Fig.3 we present the corresponding $\beta$ values, extracted from field emission measurements carried out before each spark scan. The differences in the conditioning found in the behaviour of $E_{b1}$ are reflected here by the variation of $\beta$ values as a function of the number of sparks. Indeed, the values for Mo and W remain almost always below 50 and decrease below 30 after about 20 breakdown events, whereas Cu exhibits $\beta$ values reaching 100 even after 30 sparks and no evidence for improvement is visible.

Measurements of conditioning curves were repeated at least on four sites for each material in order to verify that the behaviour was representative of the entire sample surface and the statistics of the results is shown in Fig. 4 for the breakdown field $E_{b1}$ and in Fig. 5 for the values of $\beta$. Without further calculation it appears obvious that the average value of $E_{b1}$ is higher for W than for Mo and even more so than for Cu. The difference is even larger after some breakdowns, since the lowest values for Mo and W are obtained at the beginning, whereas for Cu they are distributed all along the conditioning curve. The corresponding histograms of the $\beta$ values (Fig. 5) display a distribution with larger scattering and much higher maximum values for Cu than for the other materials, which is a further indication of the lacking of clear improvement in Cu from the point of view of the field emission. The average values ($<\beta>_G$) extracted from Gaussian fits to the histograms are listed in Table I.

After performing all the reported measurements the electrodes were inspected by scanning electron microscopy. The images at different magnifications are shown in Fig. 6 and Fig. 7 for the plane cathodes and in Fig. 8 and Fig. 9 for the rounded anode tip. Modifications of the surface topography are observed on all the samples, on both anode and cathode sites. Moreover, all the surfaces exhibit features with typical signs of melting. The modifications observed on Cu consist in narrow and deep craters. On W and Mo the modifications are extended over a larger region and the craters do not look as deep as for Cu. On the anode side (Fig. 8 and Fig. 9) the evidence for melting is demonstrated by droplet-like features, particularly on Cu and Mo. Moreover, a net of cracks forms on Mo and W (Fig. 9(b) and Fig. 9(c)), where the fractures are intergranular. It should be noted that the images of the anode surface are taken as top view, so that possible craters are less visible. No such cracks
were found in the Cu or W cathode spark zones, while micro-cracks appeared within the cathode craters or remelted zones on Mo.

4. DISCUSSION

The conditioning behaviour investigated in the dc spark test set-up can be tentatively compared with high-power 30 GHz RF measurements performed in the CTF2 (CLIC test facility 2) [11]. 56 MW of input power were needed to establish an accelerating gradient of 150 MV/m, which corresponds to a peak surface electric field of 330 MV/m and a total pulse energy of 0.8 J. The conditioning curves shown in Fig. 10 represent the achieved stable gradient as a function of the total number of RF pulses - the total number of breakdowns is about a factor of ten lower. The dc and RF results are similar in that the refractory metals consistently reach a higher surface electric field than copper, which confirms the benefits hoped by their use. However, differences are observed in the ranking of achieved gradients for the different materials: tungsten, molybdenum then copper for dc and molybdenum, tungsten and copper for RF (highest to lowest). The reasons for the inversion of ranking between tungsten and molybdenum might be an intrinsic difference between RF and dc breakdown mechanism or differences in the conditions used in the tests. Among others one could consider the different geometry, the different surface finishing of the as-received materials (grinding for the RF test and lamination for the dc test), the larger energy available for a spark in the dc system and finally the fact that RF conditioning of the tungsten iris structure was stopped due to lack of time in the test facility. The absolute values of the surface electric field limits achieved with the different materials are in the same range in the RF and dc experiments. A comparison of the limits achieved in both experiments is summarized in table I.

The relative conditioning rates of the three materials with RF is reproduced with the dc set-up, albeit with the same inversion of tungsten and molybdenum as ultimate gradient. However, the results differ dramatically in the total number of necessary breakdowns, which is much higher in the case of RF cycling. This is consistent with the assumption of a minimum necessary amount of deposited energy per surface area for conditioning. Since the available energy per breakdown event is higher in the dc system and a larger surface area is exposed to high surface electric fields in the RF tests, more breakdowns are required to condition the surface in the latter case.
In the dc test the absence of improvement for Cu is correlated with the oscillation behaviour found for the $\beta$ values (Fig. 3 and Fig. 5), which remains in average higher than for the other two materials (Table I) in the present set-up. The value of $\beta$ extracted from the Fowler-Nordheim plots depends in principle on the value chosen for $\phi$. The changes of $\phi$ for instance for Cu as a function of the exposure to O$_2$ at pressures up to 0.5 mbar are below 0.5 eV [17] and therefore the possible uncertainty on $\beta$ is of the order of 16 %, well below the difference between the values for Cu and Mo or between Cu and W. Moreover, the surface melting identified by SEM suggests that the surface, at least after a few breakdowns, is purely metallic. The correlation in the conditioning between field emission and the breakdown field confirms that field emission is crucial in order to determine the behaviour with respect to breakdown [4]. A higher $\beta$ value implies a higher local field, as shown in table 1 for the average values. The values are close to the critical breakdown fields, also defined as $\beta E$, given in Ref. [18] and in particular there is agreement on the fact that the value for Cu is higher than for W and Mo.

The observed oscillations of $\beta$ as a function of the number of sparks are a serious warning against a safe use of Cu for field above 100 MV/m. Note that in the RF experiments oscillations are not visible, since Fig. 10 shows the maximum field reached in the experiment before breakdown without specifying the breakdown rate at that field. The oscillations in the dc experiment could possibly translate in a high breakdown rate at the corresponding RF field level. Independently of the causes, any breakdown on a Cu surface will start to modify it with two main effects. First, the accelerating structure geometry will be deteriorated. Second, the accidental creation of regions of high $\beta$ will enable high field emission currents to flow, to interact with the RF field and to cause energy losses. In the case of a real accelerating structure sparks will always occur and realistic specifications are made setting a maximum finite breakdown rate [19]. In the case of Mo and W a single spark will not worsen the situation in an irreversible way, since a further conditioning can recover the performance. The reasons could be the comparatively low vapour pressure of the refractory metals at high temperature, as visible from Table II. Melting would provoke a smoothing without material loss or crater formation through catastrophic spark current enhancement due to the presence of the vapour. The smoothing should be further favoured by the high surface energy of the refractory metals (Table II).
In the past the surface energy [3] and the surface hardness [18] were often mentioned as relevant properties influencing the resistance to sparking. For instance the possibility of blunting a field emitter tip by Joule heating, thus avoiding field induced tip rupture in high electric fields, has been shown to be more likely for higher surface tension [3]. Finally, it is noted that the $\beta$ values may be associated to other causes than the high aspect ratio tips [4].

An issue, which was not considered yet and which is quite different between RF and dc, is whether the breakdown is initiated/dominated by the cathode or the anode (discussed in Ref. [3,4]). In the present investigation no clear difference was observed between the conditioning curves taken at the first site on the cathode with a virgin anode and at the further sites with the same anode, which had already suffered some breakdown events. However, electron bombardment of the anode could also play a role in the conditioning and breakdown processes. This topic will be investigated in the future for instance by using different materials for anode and cathode. Future investigations will also use lower energies stored on the capacitor to verify whether the lower available energy could enable to reach higher fields through a softer conditioning without damaging of the electrodes.

5. SUMMARY AND CONCLUSIONS

The conditioning behaviour for Cu, Mo and W has been investigated. The lowest applied breakdown field was found for Cu. Moreover, W and Mo exhibit a monotonic increase of the dc breakdown field as a function of number of sparks whereas for copper the no average improvement is found. The field enhancement factor extracted from field emission measurements has a corresponding decrease for W and Mo as a function of the number of sparks, whereas the values for Cu remain much higher. Part of the results are in agreement with the conditioning behaviour observed on the CTF2 structure. In spite of the fact that the differences and similarities between RF and dc results are not completely understood, the dc experiment gives useful indications for material supply choices and surface preparation techniques.

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### TABLES

**Table I:** Average breakdown field ($<E_{b1}>_G$), average field enhancement factor ($<\beta>_G$) and average local breakdown field ($<E_{Loc}>_G=<\beta*E_{b1}>_G$) extracted from Gaussian fits to the histograms of all conditions series. The last column gives the maximum surface field from the RF experiment [Ref].

<table>
<thead>
<tr>
<th></th>
<th>Average breakdown field $&lt;E_{b1}&gt;_G$ [MV/m]</th>
<th>$&lt;\beta&gt;_G$</th>
<th>$&lt;E_{Loc}&gt;<em>G=&lt;\beta*E</em>{b1}&gt;_G$ [MV/m]</th>
<th>Maximum surface field in RF experiment [MV/m]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>170</td>
<td>57</td>
<td>10350</td>
<td>260</td>
</tr>
<tr>
<td>Mo</td>
<td>260</td>
<td>33</td>
<td>8090</td>
<td>420</td>
</tr>
<tr>
<td>W</td>
<td>357</td>
<td>27</td>
<td>9640</td>
<td>340</td>
</tr>
</tbody>
</table>

**Table II:** List of key physical material properties possibly relevant for breakdown.

<table>
<thead>
<tr>
<th>Property</th>
<th>Cu</th>
<th>Mo</th>
<th>W</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heat of fusion [J/mm$^3$]</td>
<td>1.8</td>
<td>3.4</td>
<td>3.7</td>
</tr>
<tr>
<td>Heat of evaporation [J/mm$^3$]</td>
<td>42</td>
<td>63</td>
<td>87</td>
</tr>
<tr>
<td>$p_{\text{vap}}$ [mbar] at boiling temperature of Cu (2836 K)</td>
<td>1000</td>
<td>$1 \times 10^{-2}$</td>
<td>$1 \times 10^{-5}$</td>
</tr>
<tr>
<td>$p_{\text{vap}}$ [mbar] at melting point of W (3680 K)</td>
<td>29600</td>
<td>8</td>
<td>$5 \times 10^{-2}$</td>
</tr>
<tr>
<td>Surface free energy (surface tension) for liquid metal [mJ/m$^2$] at $T_M$ [20][21]</td>
<td>1258</td>
<td>2081</td>
<td>2596</td>
</tr>
<tr>
<td>Electrical conductivity (at room temperature) [10$^6$ Ohm$^{-1}$ cm$^{-1}$]</td>
<td>0.59</td>
<td>0.208</td>
<td>0.182</td>
</tr>
</tbody>
</table>
Figure 1 - Schematic view of the experimental setup
Figure 2 – Conditioning curve: breakdown field (applied) as a function of the number of sparks for one site on Cu (circles), Mo (squares) and W (crosses).

Figure 3 – Field enhancement factor $\beta$ as a function of the number of sparks for one site on Cu (circles), Mo (squares) and W (crosses).
Figure 4 - Histograms of the breakdown field from various conditioning curves measured at different sites of the cathode for Cu, Mo and W

Figure 5 - Histograms of the field enhancement factor $\beta$ from various conditioning curves measured at different sites of the cathode for Cu, Mo and W
Figure 6 a, b, c - Scanning electron microscope (SEM) images of the Cu (a), Mo (b) and W (c) cathodes at the position where a conditioning curve has been measured. The magnification was 250, and the tilt angle was 75 degrees. The arrow shows 100 microns.
Figure 7 a, b, c.- SEM image, magnified view (2500 times) of the regions in Fig. 5: The arrow indicates 10 microns
Figure 8 a, b, c.- Scanning electron microscope images (SEM) of the anode tips for Cu(a), Mo(b) and W(c). The magnification was 500, and the images were recorded with normal incidence. The arrow indicates 50 microns.
Figure 9 a,b,c. – SEM images, magnified view of the regions in Fig. 7 (magnification 5000). The arrow indicates 5 microns
Figure 10 - Conditioning curves with maximum surface RF field, corresponding to 2.2 times the accelerating field, as a function of number of pulses measured for CTF2 for accelerating structures of Cu, Mo and W.