Breakdown resistance of refractory metals compared to copper

M. Taborelli, S. Calatroni, M. Kildemo

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The behaviour of Mo, W and Cu with respect to electrical breakdown in ultra high vacuum has been investigated by means of a capacitor discharge method. The maximum stable electric field without breakdown and the field enhancement factor, $\beta$, have been measured between electrodes of the same material in a sphere/plane geometry for anode and cathode, respectively. The maximum stable field increases as a function of the number of breakdown events for W and Mo. In contrast, no systematic increase is observed for Cu. The highest values obtained are typically 500 MV/m for W, 350 MV/m for Mo and only 180 MV/m for Cu. This conditioning, found for the refractory metals, corresponds to a simultaneous decrease of $\beta$ and is therefore related to the field emission properties of the surface and their modification upon sparking. Accordingly, high $\beta$, values and no applicable field increase occur for Cu even after repeated breakdown. The results are compared with RF breakdown experiments [1] performed on prototype 30 GHz accelerating structures for the CLIC accelerator.

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Abstract
The behaviour of Mo, W and Cu with respect to electrical breakdown in ultra high vacuum has been investigated by means of a capacitor discharge method. The maximum stable electric field without breakdown and the field enhancement factor, $\beta$, have been measured between electrodes of the same material in a sphere/plane geometry for anode and cathode, respectively. The maximum stable field increases as a function of the number of breakdown events for W and Mo. In contrast, no systematic increase is observed for Cu. The highest values obtained are typically 500 MV/m for W, 350 MV/m for Mo and only 180 MV/m for Cu. This conditioning, found for the refractory metals, corresponds to a simultaneous decrease of $\beta$ and is therefore related to the field emission properties of the surface and their modification upon sparking. Accordingly, high $\beta$ values and no applicable field increase occur for Cu even after repeated breakdown. The results are compared with RF breakdown experiments [1] performed on prototype 30 GHz accelerating structures for the CLIC accelerator.

INTRODUCTION
The requirement of TeV energy range in linear colliders as CLIC (Compact Linear Collider) [2] implies to achieve high accelerating gradients of the order of 150 MV/m, and hence surface fields around 300 MV/m, in order to limit the machine to an acceptable length. In this electric field range extensive damages have been observed in prototype RF accelerating structures made of copper [3]. In comparison to the complex RF test facilities relatively simple instrumentation can be used to study the breakdown resistance of materials to high dc fields. We adopted this option to compare Cu with refractory metals, like Mo and W. An improvement of the maximum stable achievable field by using these refractory metals has been recently proposed [4] and obtained in prototype RF accelerating structures [1]. In spite of the fact that the mechanism leading to breakdown in dc and RF might differ in details the resistance to dc breakdown could be a prerequisite for the selection of materials for high RF field regions.

EXPERIMENTAL METHODS
The apparatus (described in details in [5]) consists of a gap junction mounted in UHV (Ultra High Vacuum, the system is baked at about 150°C) in a tip-plane geometry, where both the tip (anode) and the plane sample (cathode) are made of the same material. The tip has a hemispherical apex of 3 mm diameter. The distance between these electrodes can be adjusted in situ (at µm accuracy) by a mechanical differential lever. High voltage is applied to the anode through a capacitor. First the capacitor is charged up to a defined value by a dc high voltage source (maximum 12 kV) and then it is connected through a system of HV switches [5] to the gap junction. After 2 s the charge of the capacitor is measured by connecting it to a Coulomb-meter. Three cases are possible: i) the capacitor maintains its charge and no current flows through the gap ii) the voltage is sufficiently high to induce field emission currents and the capacitor discharges slowly iii) the voltage is high enough to induce a breakdown and the capacitor discharges abruptly. If no breakdown occurs the capacitor is charged at a higher voltage and the process is repeated until breakdown. For the case where the capacitor discharges through field emission an analytical solution has been calculated [5]. The energy available in a single pulse is 1.4 J for the capacitor charged at 10KV. From the gap width and the applied voltage the so-called first breakdown field $E_{\text{bd}}$ is calculated by assuming plane electrodes geometry. In the following the data are expressed as fields rather than voltages. By repeating such an experiment several times on the same surface site a curve of maximum stable field $E_{\text{st}}$ as a function of the number of breakdown events can be obtained. Additionally, the field emission current before each breakdown curve is measured. This is achieved by connecting the high voltage supply directly to the gap and increasing progressively the voltage; the corresponding current is monitored through an Amplifier, which is protected by a 12 MOhm resistor.

In this case the voltage is kept well below the breakdown limit and the maximum field emission current is limited at 0.5 µA. In order to characterize field emission behaviour the field enhancement factor $\beta$ is calculated by fitting the measured current $I(E)$ with the Fowler-Nordheim expression for field emission [6]:

$$I[A] = A_v \frac{1.54 \times 10^6 \beta^2 E^{-2/3} [MV/m] e^{-1.041 \phi^{-1/2}} e^{-6.53 \times 10^3 \phi^{1/2}}}{\phi e V}$$

with $\phi$ the work function and $A_v$ emitter area. For sake of simplicity the work function is taken as 4.5 eV for the three metals. Knowing the value of $\beta$ one can calculate the local field defined as: $E_{\text{loc}} = \beta \cdot E$

The samples (Cu OFE, Mo 99.9%, W 99.95%) were chemically degreased through the CERN standard procedure [7] before mounting. Four different sites were measured for each sample. After breakdown Vickers
hardness measurements were performed at load HV20 on surface sites, which were not influenced by the breakdown.

RESULTS

A single measurement of the breakdown field is shown in figure 1 for the Cu sample. The graph presents the field remaining across the gap, 2s after connection to the capacitor, as a function of the initially applied field $E_i$ (capacitor loaded). For low values of $E_i$ the corresponding $E(2s)$ is identical to $E_i$, since no current can flow. At sufficiently high $E_i$ some field emission current flows, the resulting $E(2s)$ is lower and the curve slope decreases. At sufficiently high $E_i=E_{b1}$, corresponding to about 212 MV/m in the present case, breakdown occurs and $E(2s)$ is almost zero, since the capacitor is almost fully discharged.

![Figure 1](image1.png)

Figure 1: Single measurement of breakdown field on Cu: the breakdown field is 212 MV/m. The value of the calculated [5] field emission current before breakdown is 1 mA.

Table 1: Average values from the dc tests and results from rf tests [1].

<table>
<thead>
<tr>
<th></th>
<th>Avg. $E_{b1}$ [MV/m]</th>
<th>Avg. $\beta$</th>
<th>Avg. $E_{loc}$ [MV/m]</th>
<th>Max. surface field in rf [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>

The average values for the breakdown field given in table 1 are obtained by measuring a sequence of such curves in identical conditions for the three different materials. The average has been performed on at least 50 values for each material. The ranking of the materials with respect to breakdown resistance is Cu, Mo, W, going from the worst to the best. The average field enhancement factor $\beta$ is significantly lower for W and Mo than for Cu. For the same applied field $E$ this means a lower local field $E_{loc}$ in the case of the refractory metals. The evolution of the breakdown field $E_{b1}$, as a function of the number of spark events on a single site depends on the material (figure 2). In particular for Cu there is no clear increase of the breakdown limit as a function of the number of sparks. Instead, for Mo and W the value of the maximum stable field is almost monotonically increasing (conditioning). As a consequence the average values of $E_{b1}$ for Mo and W presented in table 1, which include also initial values, are lower than those of a conditioned surface. The $\beta$ values of Mo and W remain below 45 upon sparking (figure 3), whereas for Cu they reach 100 (see also average in table 1) even after a large number of sparks. In addition they show a much larger spreading than for Mo and W (data not shown).

For all materials SEM (Secondary Electron Microscopy) reveals local melting caused by breakdown.

![Figure 2](image2.png)

Figure 2: Evolution of breakdown field as a function of the number of sparks on a single site for Cu, W and Mo.

![Figure 3](image3.png)

Figure 3: Values of $\beta$ from in field emission curves acquired before each breakdown shown in figure 2.

DISCUSSION AND CONCLUSION

The behaviour of the materials investigated with the dc spark set-up can be compared with the data (figure 4) from the high power 30GHz measurements performed on CTF2 (CLIC test facility 2) [1] where 56 MW were applied to reach 150MV/m accelerating gradient, corresponding to 330MVm surface field for a pulse
Energy of 0.8 J. Also in the rf case the refractory metals show better resistance to the electric field. However, the ranking between Mo and W is inverted with respect to dc results. This might be due either to an intrinsic difference between dc and RF breakdown mechanisms or to the difference in the material treatments or to the higher energy at disposal in the case of the dc setup. Moreover, the conditioning in the RF case was stopped for lacking of time in the test facility and the values for Mo and W in contrast to that of Cu do not show saturation. The conditioning in the case of the dc set-up needs a much lower number of breakdowns. This is consistent with the argument of a minimum energy per surface area necessary for conditioning, since in the dc set-up the energy per pulse is higher and the exposed area is smaller.

The limited value of $\beta$ and increasing value of $E_b$ as a function of breakdown events measured in dc experiments (figures 2 and 3) show that Mo and W have the advantage of recovering and avoiding irreversible deterioration after breakdown. This is not the case for Cu, which does not show a stable decrease of $\beta$ below a given limit. The comparatively low vapour pressure at high temperature of the refractory metals (table 2) with respect to Cu is likely to be the reason for the absence of important and permanent deterioration. The higher vapour pressure of Cu at comparable temperature means that it can be partly evaporated during or even before breakdown. This provokes a deterioration through material loss and possibly the vapour induces early breakdown. Notice that also for RF breakdown simulations have been performed for mechanisms based on the gas desorbed from the surface or vaporized [9] and then ionized. Moreover, for Mo and W melting can provoke surface smoothing without material loss and without catastrophic current enhancement induced by the presence of vapour. Smoothing is additionally favoured by the higher surface energy of the refractory metals (table 2) and this might be an explanation for their lower $\beta$ values. Roughness of the virgin surface is believed to be relevant, but not at the scale measured by a dynamic-focussing laser profilometer (table 2). Hardness, which is related to tensile strength, has been also quoted to be correlated with the breakdown field [10] $E_b$, but not with the maximum local field responsible of the electrostatic forces. In the present case an indication of the correlation is also found (figure 5) and should be verified on a larger number of materials.

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**Table 2: Physical quantities possibly related to the breakdown behaviour**

<table>
<thead>
<tr>
<th></th>
<th>Cu</th>
<th>Mo</th>
<th>W</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surface energy [mJ/m$^2$] at melting point [8]</td>
<td>1258</td>
<td>2081</td>
<td>2596</td>
</tr>
<tr>
<td>Pavap at 3680 K (Tm of W) [mbar]</td>
<td>29600</td>
<td>8</td>
<td>0.05</td>
</tr>
<tr>
<td>Measured Vickers hardness (dc-samples)</td>
<td>94</td>
<td>265</td>
<td>509</td>
</tr>
<tr>
<td>Roughness Ra on 4mm length (dc-samples), [$\mu$m]</td>
<td>0.43</td>
<td>0.27</td>
<td>1.55</td>
</tr>
</tbody>
</table>

In conclusion the dc tests reveal that Mo and W thanks to their low vapour pressure can withstand higher fields than Cu and even in case of breakdown they exhibit positive conditioning leading to an improvement of $\beta$ values and maximum stable field.

**REFERENCES**

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