ISBN: 978-3-95450-214-1 ISSN: 2673-5490 doi:10.18429/JACoW-IPAC2021-MOPAB362

ATOMISTIC MODELING OF THE COUPLING BETWEEN ELECTRIC FIELDS AND BULK PLASTIC DEFORMATION IN RF STRUCTURES*

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Abstract

A notable bottleneck in achieving high-gradient RF technology is dictated by the onset of RF breakdown. While the bulk mechanical properties are known to significantly affect the breakdown propensity, the underlying mechanisms coupling RF fields to bulk plastic deformation in experimentally relevant thermal and electrical loading conditions remain to be identified at the atomic scale. Here, we present the results of large-scale molecular dynamics simulations (MD) to investigate possible modes of coupling. Specifically, we consider the activation of Frank-Read sources, which leads to dislocation multiplication, under the action of bi-axial thermal stresses and surface electric-field. With the help of a charge-equilibration formalism incorporated in a classical MD model, we show that a surface electric field acting on an either preexisting or dislocation-induced surface step, can generate a long-range (r^{-1}) resolved shear stress field inside the bulk of the sample. We investigate the feedback between step growth following dislocation emission and subsequent activations of Frank-Read sources and discuss the regimes of critical length-scales and densities of dislocations, where such a mechanism could promote RF breakdown precursor formation.

INTRODUCTION

Ultra-high gradient radio-frequency (RF) structures are the engineering stepping stones to achieve compact and transportable accelerator facilities, benefitting wide-ranging application areas including bio-medical and national security challenges. However, RF-exposed surfaces in these structures become increasingly susceptible to breakdown as the operational accelerating gradients are elevated to higher intensities. Therefore, one of the key design challenges in manufacturing higher gradient accelerators is to tackle the onset of breakdown which can significantly limit the functionality of such engineering systems.

To date, several experimental observations [1-4] have revealed how different microstructural treatments (e.g., hard or annealed Copper [1]) can lead to noticeable difference in their breakdown behaviour under electric fields and thermal stresses generated due to RF losses [2]. While these experiments were successful in capturing post-breakdown signatures in severely damaged surface microstructures [2-4], the onset mechanisms of breakdown precursor formation on an initially pristine metal surface due to the presence of bulk defects, still remain poorly understood. Computational tools can provide crucial multiscale insight to the onset regime of breakdown events which is difficult to access through state-of-the art experimental capabilities. In this regard, a number of computational studies have addressed the material physics behind breakdown events mainly with the help of atomistic simulations [5, 6], mean-field stochastic modeling [7, 8] and coupled continuum-atomistic frameworks [9]. To demonstrate the formation of material instabilities under the action of dc electric fields, most of these prior studies either considered a) special subsurface microstructures [5, 6] or b) continuous coarse-grained activation of dislocations in simplified homogenized statistical mechanical models [7, 8]. General defect evolution mechanisms in bulk material microstructure, in the context of breakdown precursor formation on an otherwise clean metal surface, are yet to be explored systematically.

In this study, we consider a typical dislocation multiplication mechanism i.e., the activation of Frank-Read sources, which is widely understood to be the primary mode of plastic deformation in fcc metals at low (< 0.3 melting) temperatures. To couple the effect of electric fields with material structure at the atomic scale, we implement a charge-equilibration based formalism in our large-scale classical molecular dynamics (MD) simulations. In the following sections, we first delineate our modeling approach and then present some of the key results followed by their implications in understanding the near-breakdown physics.

COMPUTATIONAL METHOD

Charge-equilibration Molecular Dynamics

To allow for a self-consistent treatment of the charges under external fields, we implement a long-range charge equilibration (QEq) based MD simulation scheme, much in the spirit of [10]. In this scheme, the electrostatic energy of free charges is minimized under the action of electric fields. The classical electrostatic Hamiltonian of fcc EAM copper (Cu) [11] is parameterized by quantum mechanical density functional theory (DFT) datasets. This naturally results in electrostatic induction of surface charges due to electric fields, and allows for the dynamic evolution of atomic charges, allowing for a natural coupling between surface structures and electric fields.

Activation of Frank-Read Sources

Plastic evolution in fcc Cu structures is modeled by creating Frank-Read (FR) sources deep inside the material,

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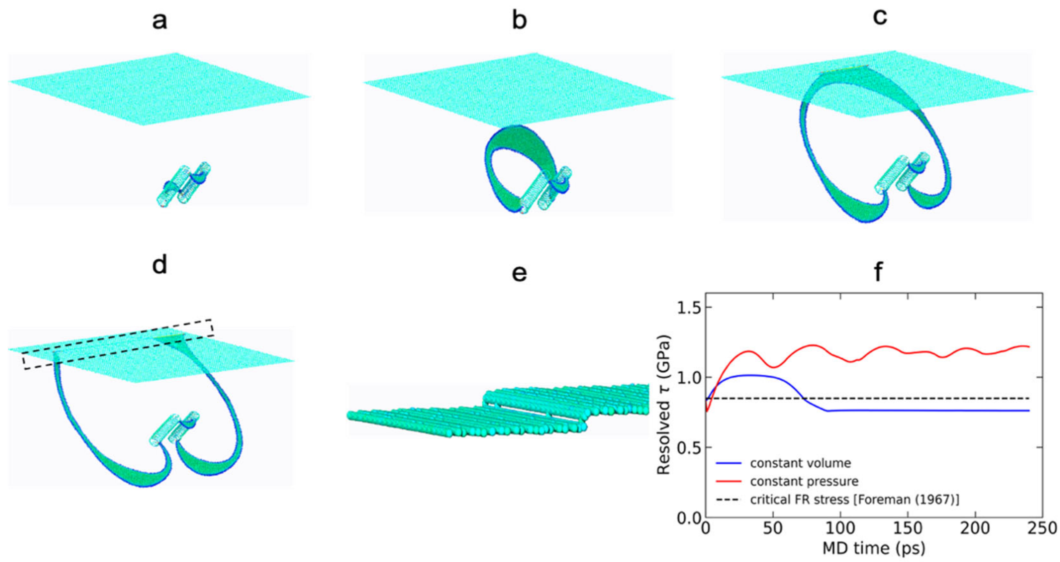

Figure 1: Activation of Frank-Read sources in fcc Cu where an initially undeformed edge dislocation in (a) starts to bow out (b) under sufficient external stress. The emitted dislocation loop then approaches the free surface (c) eventually exiting (d) to leave behind a surface step (e). Resolved shear stress required to maintain the activation (f) should be above the critical threshold (black dashed line) which might not be the case always (e.g., see the stresses for constant volume simulations depicted by the blue curve).

comprising of an edge dislocation dipole pinned by two cylindrical obstacles [12] (Fig. 1a). Through a proper choice of crystallographic orientation, these sources can be systematically activated (Fig. 1b-f) without the interference of any secondary mechanisms (e.g., cross-slip) under the action of bi-axial dilation stresses that mimics the effect of thermal loading due to RF pulse heating.

Depending on the length of the edge dislocation lines present in the FR source, a critical threshold stress [13] is required to initiate the glide along the [101](111) Cu slip plane. Under no external fields, the thermal stresses have to be just large enough to result in a resolved shear stress (RSS) which can surpass the critical threshold to maintain continuous loop emission. As an emitted dislocation loop exits the free surface and creates a slipped surface step, a drop in the RSS is expected. Indeed, as shown by our MD results in Fig. 1f, the RSS a) falls periodically as each loop exits the surface in case of our constant pressure (NPT) simulations when thermal stresses can be recovered to maintain a continuous emission, and b) drops below the threshold being unable to emit further loops when pressure is relaxed while keeping the volume constant (NVT). These two extreme cases can have significant implications during the growth of surface steps under the combined action of electric fields and thermal stresses, typical for RF accelerator structures.

RESULTS

Stress Enhancement Due to E-fields

Our charge equilibration-based model enables us to probe the effect of external electric field on the surfaces with slipped steps resulting from dislocation loop emission. An electric field of 10 GV/m strength acting normal to the surface leads to a localized stress enhancement near the step geometry shown in Fig. 2 for a ledge height of 18 b (where b is the Burger's vector magnitude which is ∼2.55 Å for Cu). Such surface steps can therefore act as damage precursors in near-breakdown conditions. We further find that a traction dipole induced by the electric field near the steps can lead to r^{-1} spatial decay of stresses inside the bulk material. This suggests that this additional contribution could activate further FR source emissions from the bulk as long as the RSS is higher than the critical activation stress at the defect location. Activation of further FR sources invariably implies the emission of more dislocation loops which can enlarge the step heights for emissions along a particular slip-plane.

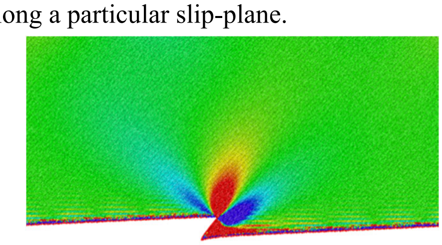

Figure 2: Localized enhancement of RSS obtained from QEq-MD simulations under 10 GV/m E-field acting normal to a surface with a step height of 18b.

Feedback Between Stress Growth Due to E-fields and Relaxation Due to Step Formation

As discussed in the previous sections, formation of a surface step through the emission of a dislocation loop near surface can relieve internal stress. This effect, when considered due to the presence of any local volume confinement, can counterbalance the enhancement effect of E-fields due to step growth. Therefore, a feedback between

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increasing RSS due to localized field enhancement and the concomitant drop due to step formation can saturate slipped step heights.

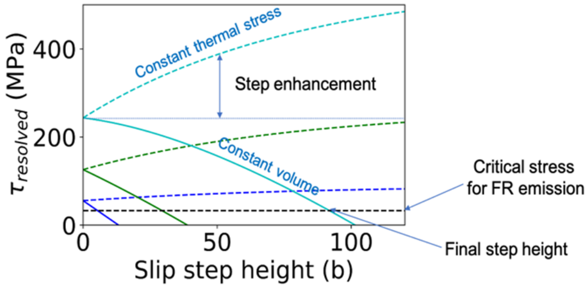

Figure 3: Model predictions for RSS evolution as surface slip step height grows under E-fields of 5 (blue), 15 (green), and 10 (cyan) GV/m and thermal stresses due to 20 °C temperature rise under the conditions described in Table 1.

Table 1: Summary of Annealed OFHC Cu Microstructural Parameters

Parameter	**Typical Value**
Dislocation density (ρ)	10^{14} m^{-2}
Distance between neighboring dislocations	1.6 µm
FR dislocation length	0.32 µm
FR activation stress	32 MPa

Under typical microstructural conditions (Table 1) in pure oxygen free high conductive (OFHC) Cu, Fig. 3 depicts two extreme cases of RSS evolution as the step height grows. If thermal stresses are uninterrupted, the RSS grows up to an asymptotic stress level, and consequently maintains a sufficiently high magnitude above the critical threshold to enable continuous loop emission, leading to uncontrolled precursor growth. However, when relaxation of thermal stresses due to surface step formation is considered, we find that the RSS starts to drop as step height increases, and eventually declines below the critical FR activation stress. This phenomenon can cease further step growth at some maximum achievable step height.

DISCUSSIONS

While we have primarily considered two idealized cases of precursor growth, in more realistic situations of multiple slip-plane activations and forest hardening of dislocations [14], the driving RSS evolution due to step growth could be anywhere between the two extreme cases considered in our study. It is important to note that the predictions in Fig. 3 are for quite strong E-fields ranging from $5-15$ GV/m which highlights the pronounced effect of step-related RSS enhancement under a nominal thermal stress level. However, under typical low E-fields, thermal stresses due to a heating temperature as low as 20 °C are sufficient to overcome the critical activation stress and to start emitting dislocation loops. This effect is promoted near breakdown when precursors formed by any other mechanism can enhance the local field.

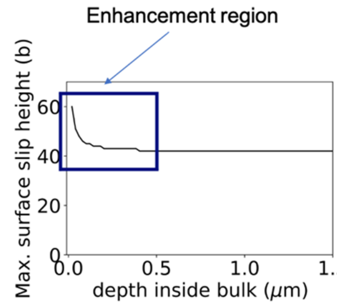

Figure 4: Maximum possible slipped step height at the surface due to emission of dislocation loops from RF sources located at different depths inside the bulk material.

To understand the individual contribution of E-field and thermal stresses in step growth, we analyze the maximum achievable surface step heights under the limiting condition of stress relaxations due to step formation. Figure 4 shows that the enhancement in step height growth is mostly due to the activation of FR sources located near the surface (~1 µm), and at strong E-fields (e.g. 10 GV/m). As an E-field of 500 MV/m can amount to a tensile stress in the order of ~ 1 MPa within an initially flat Cu surface, it is the thermal stresses which trigger the plastic deformation and surface protrusion formation due to emission of dislocations from bulk sources. Experimentally observed surface slip step formation through various electron microscopies [15] of single crystal Cu sample under purely mechanical uniaxial tensile stress tests in absence of any surface E-fields, resonates with our computational findings elucidating the importance of considering the thermo-plasticity to activate emissions under typical operating conditions of RF structures.

CONCLUSIONS

In summary, we considered general mechanisms of dislocation multiplications via Frank-Read sources in bulk fcc Cu to computationally probe the onset of breakdown precursor formation under intrinsic microstructural defect conditions, avoiding the knowledge of any special *a priori* defect structures. Our charge-equilibration based modeling framework can capture the localized enhancement of stresses due to the presence of surface steps. It is shown how plastic relaxations due to step formation can inhibit further defect growth. While strong electric fields can magnify the growth of surface steps, thermal stresses due to RF losses play the dominant role in precursor formation by activating dislocations in the bulk material promoting local enhancement of typical operating electric fields. These findings can provide crucial mechanistic insights towards achieving robust breakdown tolerant *materials-by-design*.

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