Fundamental Studies of the Surface Chemistry and Structure of Niobium Interfaces

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Overview: The technical parameters under which superconducting radio frequency (SRF) materials can be used reliably in the next generation of particle accelerators will govern many of the key design elements of these machines, arguably determining, ultimately, whether the length and costs can be brought to levels that are acceptable for accelerator production. Superconducting niobium cavities form the present technological base for such machines. The issue is whether one can manufacture, with assurance, cavities that will be able to operate at electric fields on the order of 40 MV/m. Such reliability, while occassionally achieved, has not yet been routinely achieved, with high field dissipation, field emission, and related undesirable materials related attributes forcing accelerator fields, E_{acc}, below 30 MV/m as seen in plots of the intrinsic quality factor, Q_o vs. E_{acc}. Intriguing and reproducible observations demonstrate the beneficial influence that mild heat treatments, washing with high-pressure water, and surface electro-polishing can have on the shape of the performance curve for Q_o vs. E_{acc} (hereafter referred as Q-slope improvement). These observations strongly suggest that it is the surface morphology including grain boundaries, surface cleanliness, and especially the interfacial chemistry of Nb and its associated oxides and sub-oxides (Nb_20_5, NbO_x, plus dissolved oxygen) in the interfacial region that dominate SRF materials performance. In spite of these observations, there are still fundamental physical metallurgy and interfacial chemistry issues that remain unsettled such as the oxidation kinetics of smooth and vicinal monocrystalline interfaces, the effect that heat treatment has on the composition and depth of the various metallic and oxide insulating layers that are present in the first 50 nm of the cavity, the influence of trace (or not trace) water and hydrogen on the chemistry, and, of course, the special role that grain boundaries play in this discussion.

Based on the above observations, which have come from a very broad ensemble of researchers with a focussed engineering effort on SRF materials, *the time seems particularly appropriate to revisit the fundamental surface and interfacial chemistry issues pertaining to Nb single crystals, polycrystals, alloys, and thin films deposited on such surfaces.* We herein propose such a program using our group's expertise in surface chemistry, molecular beams, thin film growth, real-time/real-space atomic-level imaging of interfaces under oxidative conditions, interfacial spectroscopy, surface work function measurements, surface alloys, and surface-phonon band structure measurements that give insight into surface forces and stability.

The deposition of surface oxygen and the mechanism for its chemical transport and reaction with the underlying interfacial region as a function of material temperature and crystalline structure appear to be crucial issues that need further exploration. *Definitive insight into the origin of the so-called "baking-effect" would represent a major achievement for this proposed program, with the potential for large impact in the the development of next generation SRF materials.*

Program Synopsis: We propose to launch a comprehensive study of the surface and near-surface oxidation chemistry of niobium single crystals (111), (110), (100), polycrystalline samples, stepped vicinal crystals, and the key alloy of this family, Nb_3Sn. We will initially seek to delineate the kinetics of the key step in this process, the transformation of surface adsorbed oxygen into domains of the Nb interfacial oxides, Nb_2O_5 and NbO_x, and the crucial "communication" of these surface and near-surface phases with the dilute solution of oxygen in the underlying Nb. How this interfacial "system" evolves as a function of temperature processing may shed light on the origin of the "baking effect" which leads to notable improvement in the Q vs. E_{acc} performance of the SRF material. Molecular-beam-based oxidation experiments coupled with *in situ* materials characterization techniques such as XPS, HREELS, STM, AFM will give new insights into the *kinetics* of the relevant oxidative transformations in the Nb monocrystals. This will complement extant work which has at least partially mapped out the statics of the chemical endpoints for the materials processing (as opposed to how those states were *kinetically* reached for a given selection of heating/oxygen dosing history). We shall also explore the role that water plays in modifying the oxidation chemistry. We also hope to explore synergistic effects in the oxidation chemistry induced by small doses of electrons, as well as the evolution of surface morphology and hence surface work function during interfacial oxidation. To summarize, we hope to add a major *fundamental* science component to the growing *engineering* effort in SRF materials at FNAL/ANL. Together, this triad of UC-FNAL-ANL based research will hopefully lead to advances in SRF materials, leading in turn to the reliable production of accelerator cavities that can operate at the extraordinary fields (ca. 40MV/m) that will be required for ILC operation.

Budgetary Resources: We propose a 3-year effort with the budget including resources for (i) personnel, (ii) supplies, materials & cryogens, and (iii) essential equipment for Year-1.

Year 1	Direct Costs (see *Note A)
1-postdoc $45k/year plus 25% benefits:	$56,250
1-graduate student, $26k/year plus U of C tuition at 50%:	$39,000
Electronics for high-resolution electron energy loss spectrometer:	$40,000
Kelvin Probe for Work Function Measurements:	$10,000
High-temp/low-temp manipulator mounts for Nb crystals:	$20,000
Supplies, Materials and Cryogens:	$34,750
Direct Costs Year 1:	**$200,000**
Year 2	
1-postdoc $45k/year plus 25% benefits:	$56,250
1-graduate student, $26k/year plus U of C tuition at 50%:	$39,000
Supplies, Materials and Cryogens:	$54,750
Direct Costs Year 2:	**$150,000**
Year 3	
1-postdoc $45k/year plus 25% benefits:	$56,250
1-graduate student, $26k/year plus U of C tuition at 50%:	$39,000
Supplies, Materials and Cryogens:	$54,750
Direct Costs Year 3:	**$150,000**
Three-Year Grand Total (Direct Costs)	**$500,000**

***Note A:** the above is a realistic 3-year budget which assumes, for the purposes of this white paper, that financial resources will come from funds that do not accrue indirect costs. If financing comes from government resources rather than UC resouces, then the appropriate UC and/or FNAL/ANL indirect cost base will obviously modify the totals and perhaps the level of effort depending on the yearly allocation. Funding levels were discussed with Dr. Lance Cooley during his visit to the Sibener Group at UC.