

R&D IN SUPERCONDUCTING RF: THIN FILM CAPABILITIES AS A GAME CHANGER FOR FUTURE SUSTAINABILITY *

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Abstract

Superconducting RF (SRF) technology for particle accelerators has been mostly based on bulk, ultrapure Nb, which has now reached its theoretical limits. Next generation technology will be based on thin films, which will be not only a way to overcome Nb performances, but also a game changer in manner of energy consumption.

This paper will describe the challenges met when trying to develop this new generation technology, and how the international community is trying to tackle it.

INTRODUCTION

Several thousands of superconductors have been identified since the discovery of superconductivity, most of them the centre of interesting, “exotic” physics, but when it comes to applications only a dozen of them are used. They are all conventional type II materials, i.e. materials exhibiting two transitions. At low field, temperature and current density, they are in the Meissner state. In presence of external magnetic field surface supercurrent generate an opposed magnetic moment and magnetic flux is fully expelled from the material. Above the first critical field H_{C1} , the external field cannot be fully screened anymore, and some flux line start to enter the material, forming vortices (a vortex defines a flux line in a normal conducting region, surrounded by screening current, while the rest of the material is still superconducting). This phase is called “mixed state”. Above the second critical field H_{C2} , the material becomes normal conducting.

Magnets vs RF Cavities

Most developments on applied superconductors have been initially devoted to electromagnets. Those materials all exhibit very low H_{C1} and very high H_{C2} and operate in the mixed state, which is desirable in this case.

In RF, vortices oscillate in the field, and moving a normal (resistive) zone gives rise to very high dissipation. So the materials optimized for magnet applications are not fitted for SRF applications; they are even the worst case scenario. Niobium is the material with the highest H_{C1} , which allows it to operate in the Meissner state. In this case, the dissipation are minimal: 10^5 less than copper at the same frequency. Superconducting cavities are the only way to reach high accelerating gradient at high duty cycles, even so in continuous wave (CW), they exhibit low field emission, no breakdown and thus reduced dark current. They also present other advantages like more open designs that help with alignment issues or reducing wake field activation...

Ultimate Limits

As SRF cavities operate in the Meissner State, in principle they cannot operate at fields higher than H_{C1} . Fortunately, when the field is parallel to the surface it is very difficult to nucleate a vortex under the surface; thus one can stabilise the Meissner state at a (superheated) field H_{SH} , above H_{C1} .

Nonetheless, stabilizing a metastable state in presence of small fluctuation is difficult. In presence of a surface defect, vortex loops can enter the material and bring an early quench (Figure 1).

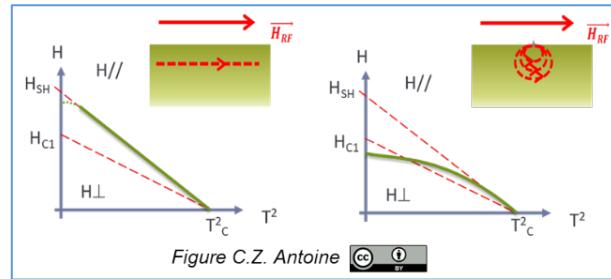


Figure 1. Ultimate field limits (i.e. transition field), in the ideal case (left) and in presence of defects (right).

So contrary to magnets, where material's optimization consist into implanting defects to enhance vortex pinning, optimising SRF material consist into reducing the density of defects on the surface and inside the material... or to find structures which are less sensitive to defects, which is more realistic in terms of industrial production.

REDUCING CRYOGENIC COSTS

The surface resistance of a superconductor follows equation (1):

$$R_S = R_0 + \frac{A\omega^2}{T} e^{-BT_c/T} \quad (1)$$

where R_0 is a small residual term, independent of the temperature T , A and B are constants, ω the RF frequency and T_c the temperature transition of the superconductor. Cavities operate between 2 and 4 K where the exponential term is small compared to R_0 . In principle, using a high T_c material should produce lower dissipation and allow operating at higher temperature. Since these materials have a smaller H_{C1} than Nb the main difficulty is to produce them in sufficiently good quality to maintain them in the superheated state.

4.5 K vs 2K: Impact on Investment Cost

Working with liquid helium rather than superfluid helium makes the cryogenic system simpler and less expensive. Pumping on helium requires a tight pumping

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system (1/cryomodules, ~250 k€), heaters for the cold gas (1/cryomodules, ~10 k€), and a specific 2K return line (~1k€/m). The cryomodules are also more complex as they have to include at minima a heat exchanger (~50 k€/cryomodule).

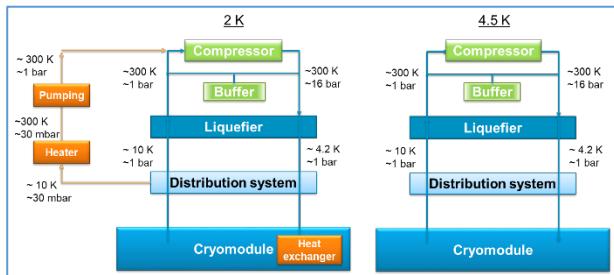


Figure 2. Main differences in cryogenic systems operating at 2K (left) and 4.5 K (right).

In Table 1, we tried to evaluate the over-costs of a 2K installation, for two examples of accelerators, based on the 2023 prices of individual components. Of course these figures are probably over-evaluated since they do not take into account cost reduction for large scale supply.

Table 1: Cost estimation of the 2 K part for two examples of recent projects.

Example	XFEL	ESS
Cryomodules #	~100	~40
Pumps	25 M€	10 M€
Heaters	1 M€	400 k€
Linac length	~1 km	400 m
Lines	1 M€	400 k€
2K total	~27 M€	~11 M€
Total cryogenic installation	80 M€ (??*)	50 M€

* For XFEL, an already existing facility was completed so the exact figures are not known

A similar evaluation was done in the 90's at LHC. The evaluation was conducted in CHF [1]. Figure 3 shows an original figure from [1] (courtesy of Philippe Lebrun), where investment cost is reported vs 1.8 K refrigeration capacity. One can observe that the 1.8 K part is 35-40% of the total cost. For comparison, pulsed XFEL, CW XFEL, and ESS necessary capacities are reported on the same graph. For instance the cryogenics for the superconducting Linac of ESS are the order of 40 M€; building a similar machine with 4.5 K capable cavities would save 14 to 15 M€ in investment.

In addition, the maintenance of the system is easier, with a lower risk of Helium pollution and pumps failure.

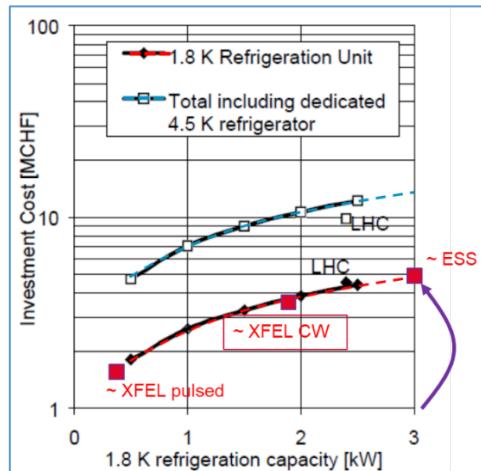


Figure 3. Investment cost in regard to the needed refrigeration power (after [1]).

4.5 K vs 2K: Impact on Operation Cost

Cryogenic costs are inversely proportional to the combination of the thermodynamic Carnot efficiency η_C and the refrigerator efficiency η_{Th} . $T_{hot} = T_{cold} / (T_{hot} - T_{cold})$. When $T_{hot} = 300$ K, $\eta_C \sim 1/70$ for $T_{cold} = 4.2$ K and $\eta_C \sim 1/150$ for $T_{cold} = 2$ K. η_{Th} is at best 25-30 % for $T = 4.2$ K and only 15-20% when $T = 2$ K.

Basically, switching from 2 K to 4.2 K divides the plug power by a factor 3. Here again, compared of ESS, supposing the Linac consumption is about 15% of the total power (250 GWh/year) [2], with 1 MWh is ~300 € (liable to increase in the future).the expected savings are also in the order of 3-5 M€. Appreciable savings can arise from changing the cavities technology to advanced thin film materials. There are still some issues to be solved: some cavity design are very sensitive to small variation of pressure that cannot be avoided in a 4.2 K system, the RF design of such cavities has to be upgraded. Moreover, the main energy budget in an accelerator is still coming from the RF power supply, which means that the savings due to cavity technology will become appreciable once RF costs have also been significantly decreased.

THIN FILMS

Today, bulk Nb cavities are not optimized for superconductivity but for thermal conduction, so that they can be stabilized in presence of tiny defects. Future relies on a functionalized material which each layer playing a different role. For instance, the mechanical and structural part can be fabricated out of copper, the surface superconducting layer can be a thin film, and one can even consider specific protective layer aimed at reducing for instance multipacting (Figure 4).

Going from a pure metal like copper to a multi-layered superconducting structure constitute a big scientific jump. Indeed copper requires just some mastering in metallurgy, a science humankind started to discover 6000 years ago. When switching to niobium, where the field penetration depth is only ~40 nm, surface features become of paramount importance. In contrast surface science is ~ one

century old. Switching from bulk to thin film materials makes surfaces and interface more even dominant. Switching to higher Tc materials requires also that the composition and structure are mastered (chemistry started only ~250 Years ago). Deposition techniques are also very recent: the development of CVD (Chemical vapour deposition) or PVD (Physical vapour deposition) started in the second part of 20th century, and the most advanced techniques are still under development... The next generation of SRF technology presents huge challenges in

material science, with a very vast parameter space to be explored.

Compared to the development of magnet technologies, the SRF R&D has been underfinanced for years. Although it is advanced material science, it is not considered a “fashionable topic” among fundamental material scientists and thus funding agencies (accelerators are thought like dirty hardware), and one must admit that material science is also treated a little like alchemy among the accelerator physicists.

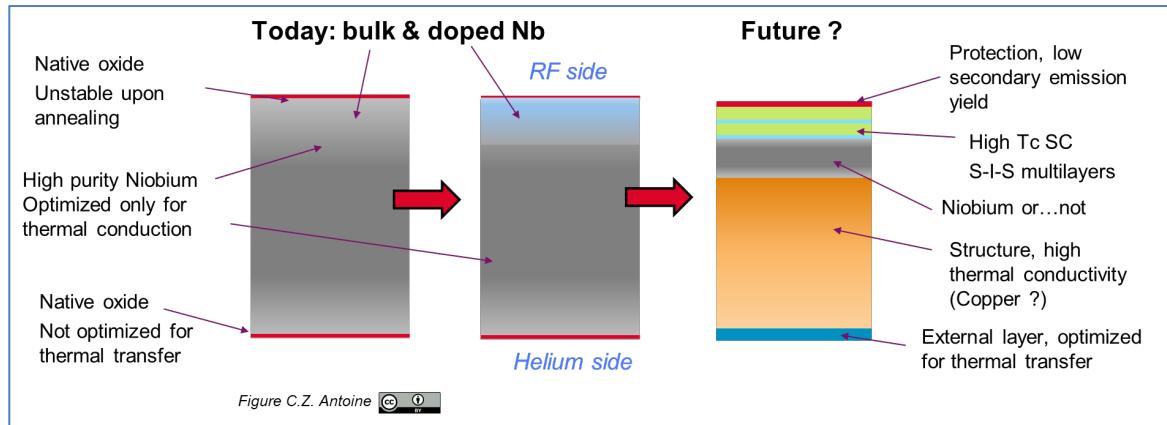


Figure 4. Possible evolution of SRF technology.

Good Thin Film Superconductor?

The best solution will not necessarily exhibit the best superconducting performances, it will rather be a compromise between:

- High superconducting and RF performance
- Easy fabrication process, high reproducibility at “industrial scale”
- Easy process to go from 1-cell to multi-cells or complex shapes
- Easy process to adapt to various frequencies
- Tunability (many of the high Tc superconductors are brittle)
- Low sensitivity to trapped flux upon cooling down
- Few crystalline defects or a structure not too sensitive to them (e.g. Superconducting-Insulating-Superconducting (SIS) structures that will be described below).

The strategy to get there is rather well established; for instance the same priorities appear in the European project IFAST WP9 [3], in the European Accelerator Road map addressed to the Laboratory Directors group by CERN [4], or the recent Snowmass 2021 Proposals [5, 6].

The common features of all these program sits on 6 priorities that will be detailed in the following chapters:

- Copper cavity production and surface preparation as a substrate for thin film deposition
- R&D on Niobium films on Copper where recent advances have been observed
- R&D of new superconductors on Cu. Up to today only Nb₃Sn cavities on Nb have been successfully tested.
- R&D on multilayer SIS structures, to gain in quality factor as well as in accelerating gradient.

- Develop new cooling systems and/or 3D printing fabrication process, aiming at reducing the needed volume of Helium and/or go to cryocooling.
- Complete material characterisation arsenal (classical techniques and specific developments)
- Increase access to RF testing (technical support, cleanroom access, RF tests access, which are most of the time monopolised by projects under construction)

Copper Substrates

If the quality of the substrate has always been paramount to the quality of the film deposition, many of the surface preparation routes that have been explored led to counter intuitive results [7]. Recently, the test of weld less structures (either bulk machined, electrodeposited, spinned...) has brought significantly progress [8, 9]. Surface preparation is also of paramount importance. The quality, especially smoothness of the substrate (Cu) appears to have a paramount importance on these performances [9-12]. Surface post treatments like laser heating are also explored [13, 14].

Well prepared Copper cavities opens the route to not only to Nb thin films, but also more advanced materials.

Most of these processes are ready for industrial transfer, which will be a necessary condition for depositing prototypes.

Niobium Films on Copper

This research field has stagnated for nearly 50 years, as many dead-ends have been explored [7], and has been limited for use in circular machine, with no need for high accelerating gradients. Indeed, the crystalline quality of these films is far less good than that of bulk Nb and the

quality factor decrease quickly with field (so called Q-slope). Nb thin film layers with performance close to bulk Nb (mitigation of the Q-slope, high transition field) were observed at CERN [15, 16] and at JLab [7]. The main features of this success are the substrate preparation (and the achievement of dense layers via energetic deposition techniques. Activities have also been conducted in Europe at UKRI, INFN, USI [11, 17, 18] and Asia at IMP and [19, 20]. CVD Deposition has been explored at Cornell [21].

New Superconductors on Cu

Nb₃Sn. Higher T_c material are expected to produce lower surface resistance. On the paper, one ideal superconductor is Nb₃Sn, with a T_c of ~18 K and H_{SH} of 425 mT (compared to ~220 mT for bulk Nb, so expected to reach accelerating gradients a factor 2 higher). Several cavities have been fabricated by thermal diffusion of Sn vapour inside Nb bulk cavities and they indeed exhibit very high Q₀ at 4.2 K [22, 23]. The ultimate field is still limited, with a behaviour indicating early field penetration at surface defects [23, 24]. There is an active R&D field to transpose these results onto copper cavities to reduce the fabrication cost of Nb₃Sn cavities. Several techniques are being explored: magnetron sputtering [25-27], electrodeposition [28, 29], bronze route [30, 31]...

The main challenge with this kind of material is to get the proper composition (the material is superconducting only for a narrow space of compositions) and the proper crystalline structure. Post-treatments like annealing are often necessary, with improvement on the crystalline quality, but detrimental side effect like Sn evaporation or Sn diffusion in the copper substrate. Alternative materials of the same A 15 family like V₃Si or Nb₃Al are also explored to overcome these problems.

The work on sample is fairly developed, but successful deposition inside cavities has not been reported yet.

NbN. Since the A15 materials are so sensitive the small composition variation, alternative materials like NbN or NbTiN¹ are also explored. They exhibit a T_c about ~17 K and a superheating field close to Nb, so they can in principle operate at 4.2 K with the same performances. They are much less sensitive than A15 to local composition, and their fabrication as thin film has been assessed for many years, since they are currently used in superconducting electronics. Active work has been conducted worldwide [5, 32, 33] Deposition set-ups for 6 GHz cavities have been designed, build and commissioned at INFN, STFC and USI [3]. First attempts of deposition in 6 GHz cavities are undergoing [34].

MgB₂. The last explored material is MgB₂, with a T_c of 39 K. Once again H_{SH} is the same order of magnitude as Nb, but it has the potential to be used at 10 K, opening the possibility to cool (small) cryomodules with cryocooler, without need of large Helium installation. The main difficulty is to find a way to deposit it inside cavity since most the discovered synthesis routes are hardly adaptable for a closed geometry.

Only few attempts have been made, mostly because of lack of financial support. An hybrid Physical-Chemical vapour deposition technique has been developed in Temple university [35], in collaboration with LANL. Promising work has been conducted on sample but deposition in a full RF cavity was never conducted up to RF test. Other techniques like magnetron sputtering or electrochemical deposition have been explored [36-39]

SIS structures. Superconducting-Insulating-Superconducting (SIS) multilayers structures were proposed in 2006 by a theoretician [40]. The first experimental proofs of concept were published in 2009 [41].

If one deposit a superconducting layer which thickness is \leq to the field penetration depth λ , it increases artificially its H_{C1} (or H_{SH}), allowing the external layer to stand higher RF field. It partially screens the field reaching the superconducting layer underneath. The use of a higher T_c materials allows to reach lower surface resistance and the presence of a dielectric layer plays two major roles: decoupling the two superconducting layers, and preventing avalanche penetration of vortex in presence of defects (see Figure 5).

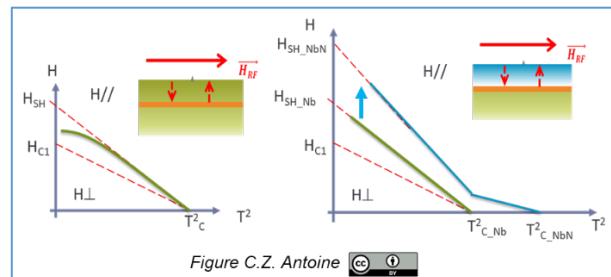


Figure C.Z. Antoine

Figure 5. Effect of a dielectric layer and multilayer concept.

With multilayers, one can both gain on the quality factor and the accelerating field. Moreover this structure is much less sensitive to the presence of defects, which are always present when fabricating complex materials. It might be the only way to produce the advanced materials mentioned earlier at large scale, with a significant yield. It opens the route to more realistic materials. Promising results have been achieved on sample in several Labs [7, 25, 42].

Material characterization

The development of complex material requires thorough characterization tools. Classical material characterization: optical and confocal microscopy, SEM, EDX and EBSD, Ion beam miller for cross-section, X-Ray, TEM, and basic superconducting properties are measured: T_c, RRR, DC magnetometry, AC susceptibility.

In addition, specific original characterization tools have been developed to measure the superconducting samples behaviour in condition closest to the operating cavities condition: Tunnelling microscopy (Superconducting gap, density of superconducting states cartography), flux penetration measurement set-ups.

¹ The presence of Ti just helps to stabilize the correct crystalline phase.

Now that the R&D community is at the eve of depositing the first prototypes, access to SRF testing hardware (surface preparation, cleanrooms, RF test stands...) will be necessary at larger scale, which will be probably soon become a bottleneck.

CONCLUSION

Thin superconducting films are liable to lead to appreciable cost savings in accelerator technology, from the investment point of view as well as operation costs. Only RF test of several prototypes will allow to determine the best material since several requirements need to be met, mainly RF, superconducting properties but also fabrication ease and yield, tunability, etc.

Substantial support is still needed if one which to apply this technology in the near future.

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