A15 SUPERCONDUCTORS: AN ALTERNATIVE TO NIOBIUM FOR RF CAVITIES

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Abstract

Since the International Committee for Future Accelerators recommended that the Linear Collider design should be based on the superconducting technology, the scientific world interest is now focused on further developments of new resonant cavities fabrication technics and cost reduction.

Besides the attempt to improve the Nb sputtered on Cu accelerating structures performances, it is important to pursue research on new materials. The goal will be the achievement of superconducting cavities working better than the Nb ones at 4.2K. Among the possible candidates, A15 compounds appear to be the most promising.

Three of them were chosen: Mo-Re system which shows the A15 phase far from the stoichiometric composition (Mo_3Re), V_3Si that has a really high RRR value, Nb_3Sn that is the only A15 compound already used for a resonant accelerating structure [1] and we obtained some interesting preliminary results.

We are setting up a 6GHz cavities measurement system: it is very simple, fast to use and it will give us the opportunity to make such small resonators become our samples.

THE BCS SURFACE RESISTANCE

The BCS surface resistance can be written as follows:

$$R_{BCS} \cong \frac{R_n}{\sqrt{2}}\left(\frac{\hbar\omega}{\pi\Delta}\right)^{\frac{3}{2}}\frac{\sigma_1}{\sigma_n} = A\sqrt{\rho_n}\,\frac{e^{-\frac{\Delta}{K_B T}}}{\sqrt{sT_c}\,T\left(1+e^{-\frac{\Delta}{K_B T_c}}\right)^2}\,\omega^2 \ln\frac{\Delta}{\hbar\omega}$$

with $A=6.0\times10^{-21}[\Omega K^2/ms^4]^{1/2}$

It's dependence on ρ_n and T_c represents an immediate criterion for selecting the most favourable candidates for cavities [2]. Unfortunately the BCS surface resistance at T=0 in practice never vanishes because of a temperature independent residual term (several nΩ). Both "physical phenomena" and "accidental mechanisms" contribute to parasitic losses so it is impossible to give one formula predicting them. Nevertheless we can deduce some interesting information from the literature analysis: R_{res} increases with R_n (the surface resistance in the normal state) by a law not weaker than proportionality. R_{res} is at least proportional to the square root of ρ_n. Between two different materials with different values of T_c and ρ_n, having the same R_{BCS}, the one with lower ρ_n should have the smallest R_{res}.

Among some fifty A15 compounds, a few could have a practical interest for RF applications: Nb_3Sn, Nb_3Al, Nb_3Ga, Nb_3Ge, V_3Ga, V_3Si, the Mo-Re system.

A15 MATERIALS

The A15 materials structure is classified as the W_3O or Cr_3Si type and is sketched in figure 1.

The stoichiometric composition is A_3B: A is a transition metal, B can be a transition metal or not. A atoms arrange themselves in rows parallel to the three crystallographic directions [100], [010], [001].

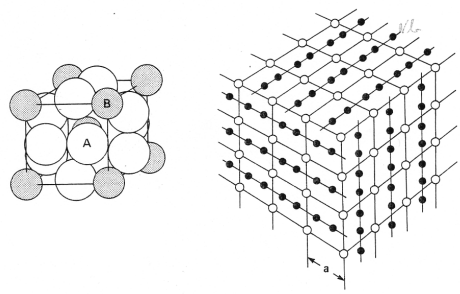

Figure 1: A15 material structure [3]

T_cs of most A15 compounds are strongly influenced by the Long-Range crystallographic Order (LRO) degree, especially when the B atom is not a transition metal [4].

Mo-Re SYSTEM

In the past, this material has been studied a lot by different scientists and it is easy to deduce some interesting informations from the literature. The main reasons of such a material choice are summarized below.

- A $Mo_{100-x}Re_x$ film superconducting critical temperature is usually higher than the bulk sample one, whatever technique is used for its deposition [5]. The coating T_c value depends on the composition and it reaches its maximum (about 15 K) for x ranging between 25 and 62 [6,7]. This thin film critical temperature increase is related to the presence of the

A15 metastable phase [5,7] (it doesn't appear in the bulk Mo-Re system phase diagram [8]).

- The maximum T_c of 15 K is achievable if the deposition temperature is higher than 1000°C: the necessary substrate temperature depends on the target composition. Nevertheless, superconducting critical temperatures of above 11 K are reported for depositions performed at about 300 °C [9].
- Bulk $Mo_{75}Re_{25}$ has already been applied for cavity prototyping [10].
- The solubility of interstitial elements, particularly oxygen, is low in Mo-Re alloys and there is indication that such contaminations do not depress superconductivity.
- If compared to other materials, Mo-Re shows a low value of the GL parameter and consequently rather high H_{c1} and coherence length. This contributes to decrease the effect of small inhomogeneities by the proximity effect.

We decided to use the single target magnetron sputtering technique choosing three different compositions: $Mo_{75}Re_{25}$, $Mo_{60}Re_{40}$, $Mo_{38}Re_{62}$. A high substrate temperature was maintained and we tried to improve the film quality with an annealing treatment: our preliminary results are encouraging.

$Mo_{75}Re_{25}$

The first depositions have been made on sapphire substrates holding them at a temperature between 600 and 1000°C. A 2" single target and a substrate heater, essentially made of an inconel insulated wire in a SiC matrix, were used.

In some cases, at the end of the sputtering process, the coating annealing treatment was performed: it has been made at the deposition temperature. The annealed films we measured show very sharp superconductive transitions ($\Delta T_c \sim 0.01$ K): we deduce this procedure gives us the opportunity to improve our films homogeneity. An annealed film superconducting transition curve is reported in the following figure.

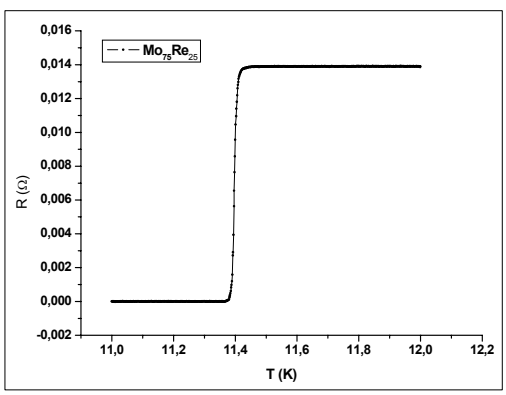

Figure 2: An annealed sample superconducting transition curve: deposition T = 633°C, annealing time = 15 min, T_c = 11,40, ΔT_c = 0.01 K, RRR = 1.57

All the T_c, ΔT_c and RRR measurements have been performed using the four contacts method.

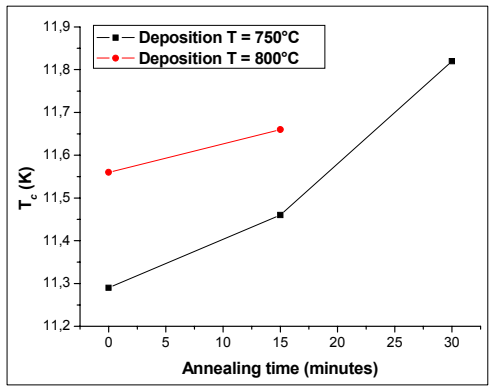

Figure 3: T_c vs annealing t at different deposition Ts

Analysing figure 3, it is possible to deduce a T_c increasing while rising the deposition temperature. Moreover when the annealing time is longer, the film T_c is higher.

Looking at the possible RF application of this material, we tried to make some depositions on copper and niobium too. A transition curve exemplum is sketched below.

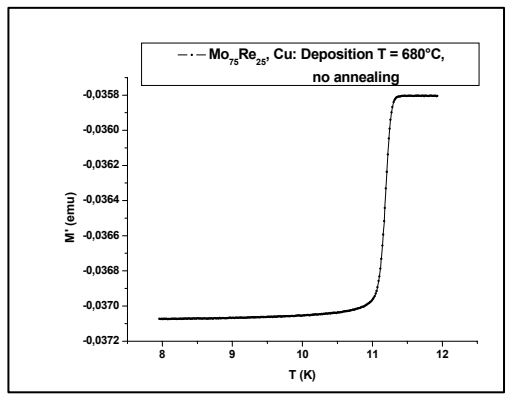

Figure 4: A $Mo_{75}Re_{25}$ film deposited on Cu transition curve: deposition T = 680°C, T_c = 11.18, ΔT_c = 0.08 K.

We just report the data obtained for a $Mo_{75}Re_{25}$ film deposited on Nb at T = 725°C: its superconductive transition temperature is 11.03 K, while ΔT_c is 0.08 K. Here we performed inductive measurements.

$Mo_{60}Re_{40}$

All the depositions have been made on sapphire substrates holding them at a temperature between 600 and 1000°C. The superconducting properties have been investigated trough the standard DC four probe method.

A superconducting transition curve example is reported: here the target composition is $Mo_{60}Re_{40}$. Knowing its deposition T is 750°C and the chosen annealing time is 60 min, a T_c of 12.00 K, a ΔT_c value of 0.041 K and an RRR equal to 1.88 were measured.

We decided to monitor how ΔT_c changes while increasing the annealing time at different deposition temperatures. Looking at figure number 5 it is easy to understand there is a ΔT_c progressive reduction.

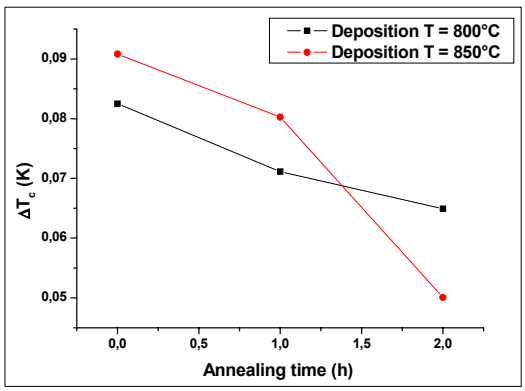

Figure 5: ΔT_c vs annealing t at different deposition Ts

$Mo_{38}Re_{62}$

We just report a superconducting transition curve example when the target composition is $Mo_{38}Re_{62}$. The superconducting properties of the sample deposited on sapphire at 750°C and annealed at the same temperature are: T_c 9.47 K, ΔT_c 0.029 K and RRR 1.11 (standard four probe DC resistivity measurement).

The maximum T_c values, obtained for films characterized by different compositions, are reported in the following table.

Table 1. The maximum T_cs

	T_c	ΔT_c	RRR	$T_{substrate}$	annealing T	annealing t
$Mo_{75}Re_{25}$	11.82K	0.012K	1.71	751°C	751-793°C	30 min
$Mo_{38}Re_{62}$	9.47K	0.029K	1.11	750°C	750-785°C	60 min
$Mo_{60}Re_{40}$	12.13K	0.065K	1.3	800°C	800-856°C	60 min

$Mo_{38}Re_{62}$ coatings T_c values are always quite low: the maximum T_c has been reached with the 40 at.% of Re.

T_c is not the only parameter that makes a compound ideal for RF applications: it is important to consider its resistivity in the normal state, just before the superconducting transition too.

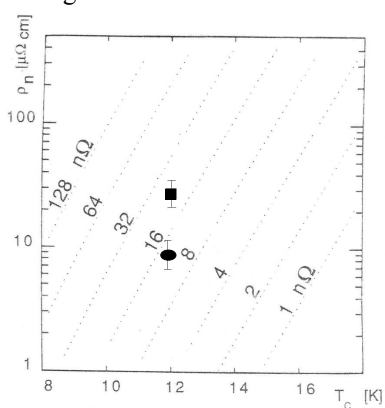

Figure 6: $Mo_{60}Re_{40}$: T_c = 12.13 K, RRR = 1.3, ρ_n ~ 30 μΩcm; $Mo_{75}Re_{25}$: T_c = 11.82 K, RRR = 1.71, ρ_n ~ 10 μΩcm.

Since R_{BCS} is a function of ρ_n and T_c, the nomogram of figure 6 can be constructed for materials at 4.2 K, 500 MHz and a strong coupling factor of 4. For a certain value of R_{BCS}, on the corresponding curve there are all the materials with different T_cs and ρ_ns having the same R_{BCS} [2].

R_{BCS} is around 16 nΩ for the films of both the compositions: $Mo_{60}Re_{40}$ and $Mo_{75}Re_{25}$.

V_3Si

The A15 phase forms congruently from the melt and it is stable between 19 and 25 at.% silicon. T_c increases linearly while reaching the stoichiometric composition so the maximum critical temperature (17.1K) is achieved with the 25 at.% of silicon. The variation of resistivity in normal state before transition has been studied in detail: at the stoichiometric composition RRR values of 80 are achievable.

The first attempt to obtain superconducting V_3Si films has been made years ago using the cosputtering method (facing target magnetron). The substrate temperature has always been maintained higher than 500°C: the films silicon content was between 21 and 32at.%. A V-Si coating composition was related to the two targets sputtering voltages. The best sample got an RRR 4.5 and its T_c value was 14.4 K [11].

The reactive sputtering of V-Si films by a DC facing target magnetron configuration in Silane/Argon atmosphere has been investigated too. The process temperature strongly influences the film composition as well as its quality. Stoichiometric superconducting V_3Si coatings have been obtained under different silane partial pressures and film deposition rates. The best samples were sputtered at 500°C at a low deposition rate and then annealed at 800°C in SiH_4 atmosphere[12].

In the light of the latest experiments V_3Si samples were grown by Si thermal diffusion into vanadium films. V coatings have been deposited onto four different substrates: sapphire, niobium, vanadium and copper [13].

The superconducting features of diffused V_3Si films on sapphire are good: T_c is higher than 16.8 K, close to the bulk value. Because of the large diffusion rate of V in Nb, no superconducting phase was produced when silanizing vanadium films onto niobium substrates. Bulk vanadium annealed in silane atmosphere gives good results. Knowing that the thermal diffusion is performed at 800°C, when they used a Cu substrate they sputtered vanadium onto both sides of the sample in order to prevent copper evaporation during the process.

This work will be pushed ahead changing the samples process temperature, silanization time and annealing time in vacuum. Our goal will be the V_3Si 6GHz cavities production and their fast measurements to optimize all the parameters for the next best recipe application to 1.3 and 1.5 GHz cavities.

Nb_3Sn

Up to now Nb_3Sn is the only A15 material that gave promising results when applied to the construction of a real cavity [1]: Sn was reactively diffused into bulk Nb cavities (1.5 GHz single cell cavity: Q factor ~10^{11} at 2 K, accelerating field ~12 MV/m).

According to the Niobium-Tin phase diagram, the A15 phase is unstable below 775°C. Nb_3Sn is the only stable compound at a temperature higher than 930°C while several spurious phases are possible at a lower T value.

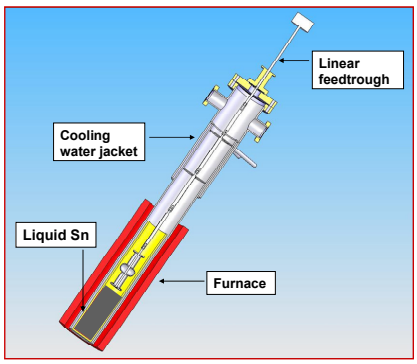

Figure 7: A schematic view of our system vacuum chamber

Thermal diffusion is a promising technique, having been already proved by the work on Nb_3Sn at Wuppertal [1]. They used the vapour diffusion technique: we decided to modify it performing the liquid solute method. It is based on a process used for cables production.

The built system is sketched in figure 7: trough the linear feedtrough it is possible to move the sample from the cold zone to the tin bath and vice versa (the liquid metal is held inside an alumina tube). Even if a 6GHz cavity is drown in the figure, we have just introduced small Nb pieces in the chamber (~20×10×3 mm^3). To avoid the possible contamination due to the presence of different elements, everything inside the vacuum system is made of niobium. The only exception is a Ti shield we decided to position in the cold zone: it reduces the heat dispersion and behaves as a good oxygen getter. The process temperature is measured trough a chomel-alumel thermocouple fixed between the chamber external wall and the alumina tube containing liquid tin. It is possible to reach a temperature higher than 930°C with an external furnace which wraps the chamber bottom part.

After fixing the sample to the Nb rod with a niobium wire, we start pumping the vacuum system. We usually bake the chamber bottom part using the external furnace (~150°C) to reach a base pressure of 10^{-8} mbar. Then the temperature is increased: as mentioned above it is important to guarantee the bath T is higher than 930°C. The following step consists in the substrate dipping inside liquid tin, then the sample is maintained in the hot zone, immediately above the Sn bath, for some hours. At the end of the process the linear feedtrough is lifted up and the furnace removed to avoid the eventually formation of Nb-Sn spurious phases.

Figure 8 shows a Nb_3Sn sample SEM image: the presence of the A15 material film is evident. This sample has been lapped and chemically etched (1:1:2 for 30 seconds) before it was analysed.

We measured the surface layer composition (EDS) and we could confirm it is not far from the stoichiometric one (figure 9).

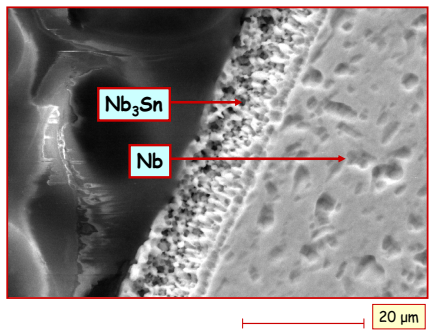

Figure 8: A SEM image of a Nb_3Sn sample. It has been dipped for 30 minutes and maintained in the hot zone for 6 hours. The measured T is 970°C.

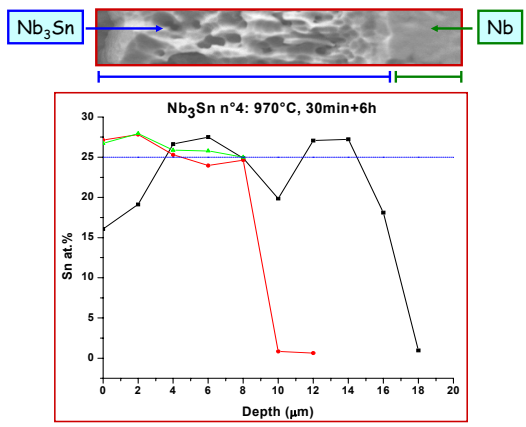

Figure 9: The surface layer composition (EDS)

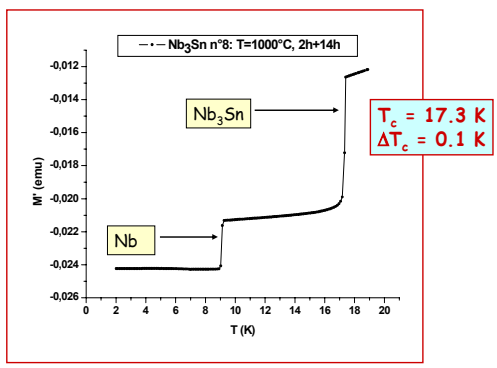

Figure 10: A Nb_3Sn sample superconductive transition curve

The best sample was obtained at 1000°C (figure 10): it has been maintained 2 hours inside the Sn bath and 14 hours in the annealing zone (in the presence of tin vapours). Its T_c is 17.3 K and ΔT_c is 0.1 K (inductive measurement).
Decreasing T until 2 K the presence of other superconducting transitions (between 2 and 9 K) was not observed so it is possible to say there are not different Nb-Sn phases.

6GHz CAVITIES MEASUREMENT SYSTEM

A large amount of samples can be produced but it is difficult to measure their RF resistance.
6GHz seamless cavities obtained by the spinning technique represent the solution: they are made from larger cavities fabrication remaining material, they don't need welding (even for flanges) and finally they can be directly measured inside a liquid helium dewar as shown in figure 11.

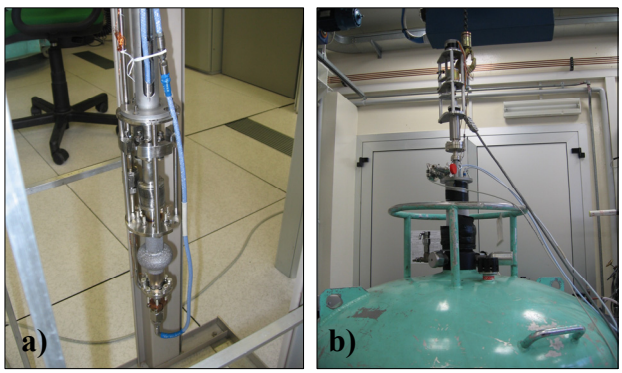

Figure 11: a) A 6GHz cavity flanged to the measurement system, b) the measurement system inserted in the He dewar.

The new 6 GHz cavities measurement system is very simple, fast to use and it will give us the opportunity to make such small resonators become our samples.

CONCLUSIONS

Using the magnetron sputtering technique we deposited many Mo-Re films and the superconducting critical temperature is higher than 12 K ($Mo_{60}Re_{40}$). The annealing treatments seem to give surprisingly good results: comparing an annealed sample to a film obtained reducing immediately the substrate heater temperature, we measured a higher T_c and a lower ΔT_c. R_{BCS}, calculated trough the nomogram is around 16 nΩ ($Mo_{75}Re_{25}$, $Mo_{60}Re_{40}$).

Looking at the V_3Si coatings preliminary results, it doesn't matter how good the initial superconducting properties of the film are: T_cs of 17 K and RRR values of 18 can be recovered by an annealing process at high temperature in SiH_4 atmosphere. Now we are ready to apply the thermal diffusion method to 6 GHz cavities. For Nb_3Sn we decided to use the liquid solute diffusion trying to improve the Wuppertal method.

Liquid tin wets the Nb surface and diffusion starts: no nucleation sites on the substrate surface are required and the Nb_3Sn layer grows very fast. The A15 film uniformity is ensured and the stoichiometry is maintained. It is possible to avoid Nb-Sn low T_c phases formations leaving T > 930°C during the experiment and reducing T very fast at the end of the process. A possible Sn outer layer has to be removed: we are able to get rid of it by a prolonged post annealing.

Figure 12: 6GHz cavities made of Nb (in the upper part) and V (in the bottom part).

From scrap material and by a seamless technique we are planning a 6 GHz cavities mass production to investigate A15 intermetallic compounds.

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