MAGNETIZATION AND SUSCEPTIBILITY MEASUREMENTS ON NIOBIUM SAMPLES FOR CAVITY PRODUCTION

M. BAHTE, F. HERRMANN, P. SCHMÜSER

II. INSTITUT FÜR EXPERIMENTALPHYSIK, UNIVERSITÄT HAMBURG
AND
DEUTSCHES ELEKTRONEN-SYNCHROTON (DESY)
NOTKESTRASSE 85, 22603 HAMBURG, GERMANY

I. Introduction

The high design gradient of 25 MV/m in the TESLA cavities puts demanding requirements on the quality of the niobium sheets used for cavity production and also on the cavity preparation steps. Part of the quality control procedures are magnetization and ac susceptibility measurements on niobium samples which accompany the cavities in all stages of chemical etching and heat treatment. An apparatus has been built that permits magnetization hysteresis measurements at 4.2 K in a dc field of up to 0.5 T parallel to the niobium surface, using the "moving sample" technique. In the same cryostat the attenuation of a small ac magnetic field (amplitude typically 0.01-0.3 mT, frequency around 175 Hz, orientation perpendicular to the niobium surface) can be determined as a function of a dc magnetic field of up to 2 T. The first experiment yields the amount of magnetic flux pinning, mainly in the bulk niobium, while the second is a sensitive measure of the upper critical magnetic field at the RF surface of the niobium sample. Experimental results are presented on niobium samples from two manufacturers at different steps of thermal and chemical treatment.

II. Experimental Procedure

A. Sample Preparation

The high purity niobium sheets for the cavity production were manufactured by Heraeus with a residual resistivity ratio (RRR) of about 270 and by Wah Chang with an RRR of about 350. The investigations were carried out on small samples with a size of $9*9*2.8\ mm^3$, cut out of the sheets by electroerosion. Alternatively, cutting with high-pressure water caused little damage of the crystal structure while conventional machining

preparation status	I. untreated	II. chemically etched	III. annealed at 800 °C	IV. heat treat-ment at 1400 °C	V. chemically etched
$RRR_{Wah\ Chang}$	350	370	475	700	700
$RRR_{Heraeus}$	270	210	250	380	380

Table 1: Residual resistivity ratios (RRR) of the Wah Chang and Heraeus niobium samples after the consecutive preparation steps. The RRRs have been measured by the Eddy current method.

led to significant plastic deformation. After cutting the samples were treated similar to the cavity preparation with the following consecutive steps:

I. Cleaning in an ultrasonic bath with ultrapure water with a detergent in order to remove contamination like oil and dust.

II. Chemical etching (buffered chemical polishing: BCP) of the surface. The etching was done by a mixture of hydrofluoric acid, nitric acid, and phosphoric acid with a ratio of 1:1:2. The amount of etching was about 70 µm or 170 µm. An alternative preparation method was the grinding of the sample surface with silicon carbide abrasive paper. Furthermore, electropolishing has been tested.

III. Annealing at 800 °C for 2 hrs under ultrahigh vacuum (UHV) conditions.

IV. UHV heat treatment of the niobium samples at 1400 °C for 1 hr and at 1250 °C for 3 hrs. The samples were surrounded with titanium sheets to deposit a thin titanium layer onto the niobium surface for gettering impurities out of the niobium bulk.

V. Chemical etching of about 100 µm in order to remove the titanium layer from the surface.

The relating RRR values after the specific steps are given in Tab. 1.

B. Cryostat

The magnetization and susceptibility measurements have been carried out at a temperature of 4.2 K in a self-built cryostat shown in Fig. 1.

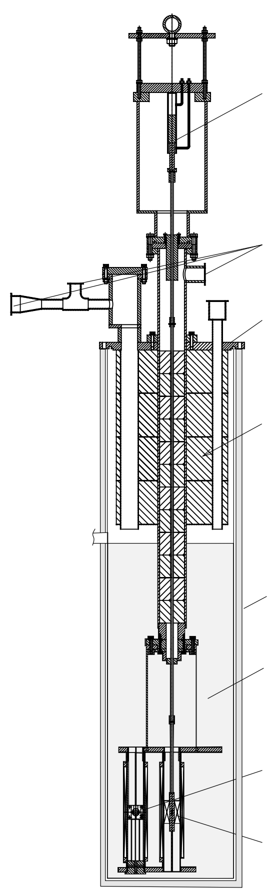

Figure 1: Cryostat with facilities for magnetization and susceptibility measurements at 4.2 K.

C. Magnetization Measurements

The magnetization measurements have been done in dependence of a uniform dc magnetic field by moving the samples through two pickup coils of opposite winding sense as shown in Fig. 2 following the suggestion of M. Wake et al. [1]. The external field was provided by a superconducting solenoid with a maximum field of 500 mT and a field inhomogenity less than 1 % over the distance swept by the samples. The sample magnetization caused by this external field was measured by a 300 step integration of the induced pickup voltage. The movement of the sample with a constant velocity took place by an electropneumatic cylinder.

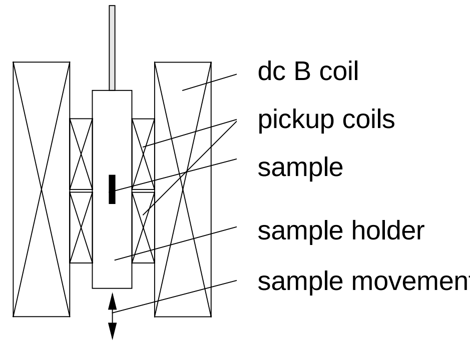

Figure 2: Schematic sketch of the magnetization measurement.

D. Susceptibility Measurements

The susceptibility measurements were performed by applying an alternating magnetic field perpendicular to the sample surface and detecting the transmission of the magnetic flux by a pickup coil at the opposite side of the sample as shown in Fig. 3. This was done as a function of an external dc magnetic field of up to 2 T parallel to the niobium surface. The ac field usually had an amplitude of 10 µT and a frequency of 175 Hz. The induced voltage in the pickup coil was analysed by a lock-in amplifier, which gave a signal proportional to the complex susceptibility of the sample. This measurement method has been proposed by Weingarten et al. [2] [3].

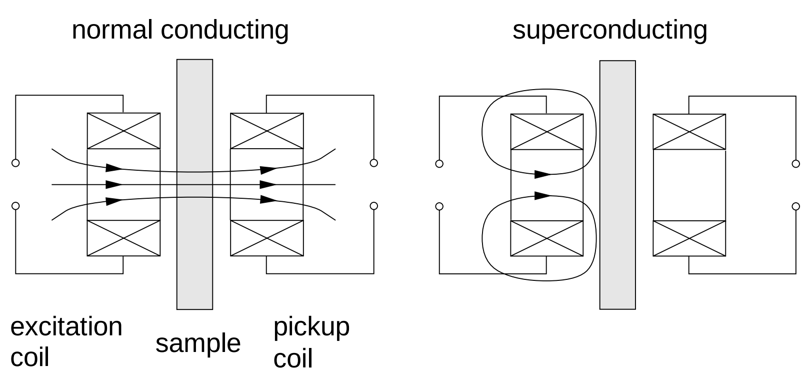

Figure 3: Schematic sketch of the susceptibility measurement. The external dc magnetic field is applied parallel to the sample surface.

III. Results and Discussion

A. Magnetization Measurements

Typical virgin curves of the Wah Chang and the Heraeus material after the different consecutive preparations (untreated, chemical polishing, annealed at 800 °C, high temperature heat treatment at 1400 °C with titanium getter, chemical polishing) are shown in Fig. 4. Visible is the linear increase of the magnetization with increasing dc magnetic field resulting from the ideal diamagnetic behavior (Meissner-Ochsenfeld effect). After reaching a maximum the magnetization drops as magnetic flux starts to penetrate the bulk until the upper critical magnetic field B_{c2} of ≈ 270 mT (4.2 K) is reached. The B_{c2} value is independent of the sample preparation. Beyond B_{c2} the metal is in the normal conducting state.

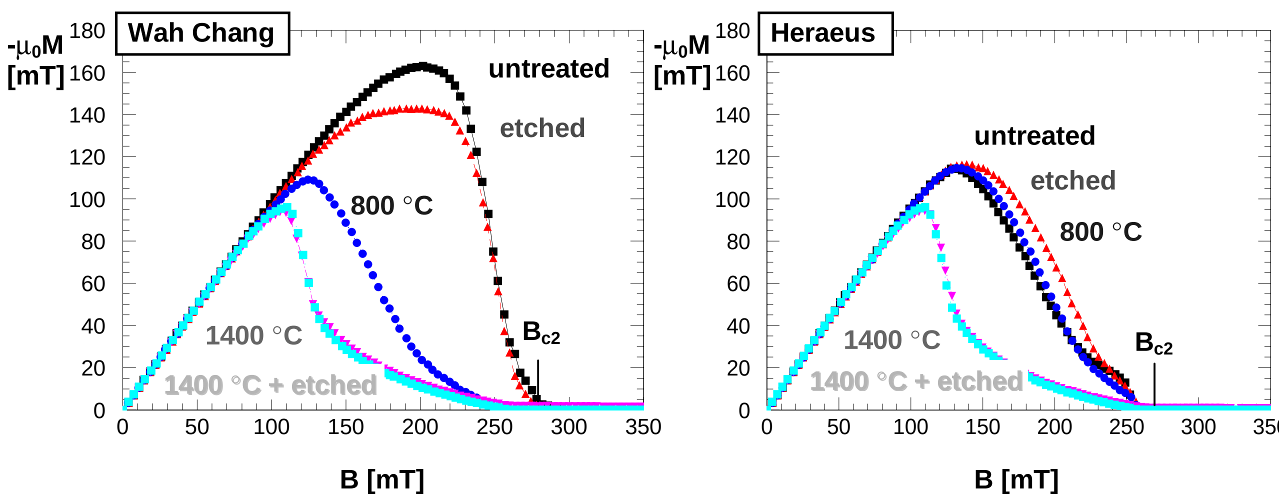

Figure 4: Virgin curves of the magnetization measurements of the Wah Chang (left) and Heraeus (right) material untreated and after different consecutive preparation steps (BCP, annealing at 800 °C, heat treatment at 1400 °C plus titanium getter, BCP).

For an ideal type II superconductor, the lower critical magnetic field B_{c1} is given by the maximum of the magnetization curve. From Fig. 4 it is obvious that the characteristic type II behavior is approximately observed after the 1400 °C heat treatment but not in the previous preparation steps. The critical field B_{c1} can be derived from the magnetic field at maximum magnetization, however the influence of flux pinning as well as demagnetization effects due to the geometry of the samples have to be taken into consideration.

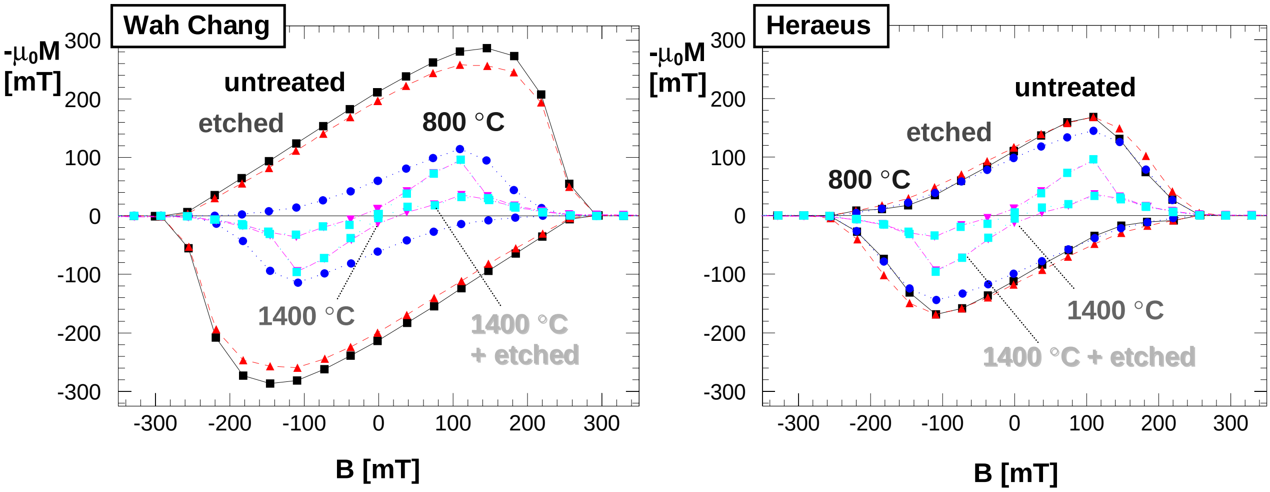

Figure 5: Magnetization hysteresis of the Wah Chang (left) and Heraeus (right) material untreated and after different consecutive preparation steps (BCP, annealing at 800 °C, heat treatment at 1400 °C plus titanium getter, BCP).

The amount of flux pinning can be inferred from the hysteresis curves in Fig. 5. The hysteresis area and the remaining magnetization at vanishing external field are both decreased during the consecutive preparation steps. After the first BCP a slight reduction is observed for the Heraeus as well as for the Wah Chang samples, explainable by the removal of the surface damage layer caused by fabrication and thus removal of pinning positions [4]. By the 800 °C annealing the Wah Chang samples exhibit a strong decrease of the hysteresis area while the hysteresis of the Heraeus material remains nearly unaffected. This behavior can be explained by an insufficient final annealing of the Wah Chang niobium sheets resulting in smaller grain sizes and therefore increased number of grain boundaries acting as pinning centers. This is visible in the micrographs of the chemically etched niobium surfaces shown in Fig. 6. The Wah Chang material shows crystallites with average diameters of about 25 μm in contrast to the Heraeus sheets which have been annealed at the company above the recrystallization temperature of about 750 °C yielding crystallite sizes in the range of 75 μm. Remarkable is also the decreased RRR of the Heraeus samples from 270 down to 210. An explanation for the decrease might be the input of hydrogen into the bulk material during the etching process. The Wah Chang material which was surface-hardened by skin rolling does not show this loss in RRR. During the 800 °C heat treatment lattice defects are annealed and recrystallization takes place. Thereafter the grain sizes are comparable and the two different materials show an equivalent amount of flux pinning i. e. a homogenization [5]. By the heating also hydrogen is removed from the bulk. The RRR value of the Heraeus material is again increased up to 250. The slightly smaller zero-field magnetization and hysteresis area of the Wah Chang material may be caused by a lower impurity content which can be inferred from the higher RRR of 475.

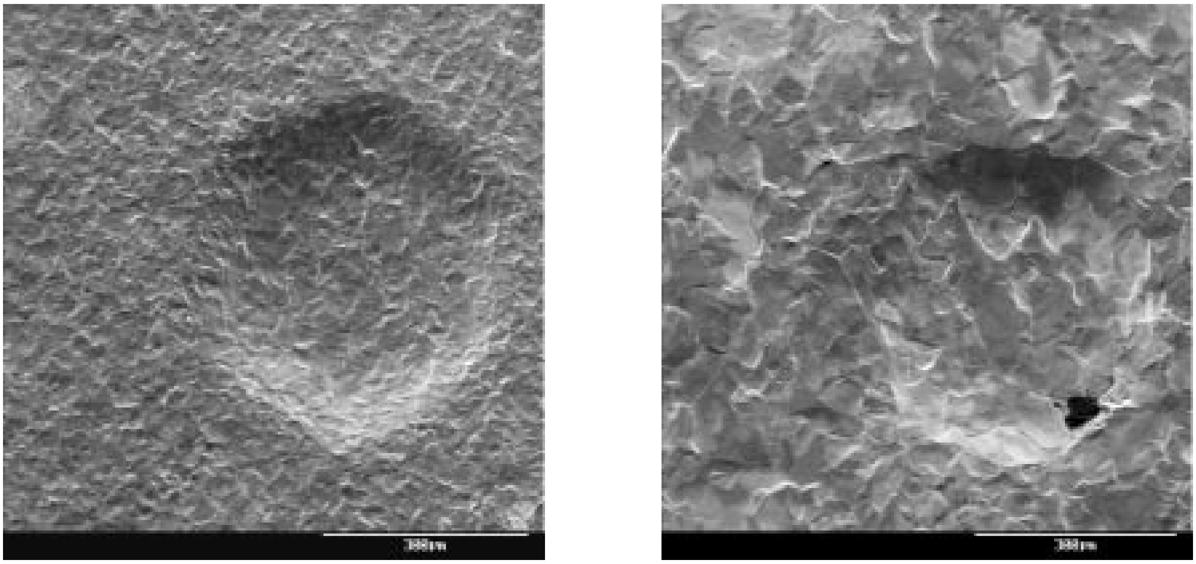

Figure 6: Micrographs of the Wah Chang (left) and the Heraeus (right) surfaces after BCP of 80 μm made by a scanning electron microscope. The impressions have been done by a diamond tip for a purpose not presented here.

Further heat treatment at 1400 °C together with titanium getter reduces the amount of dislocations and impurities, especially oxygen and nitrogen, and leads in addition to a considerable growth of the niobium grains up to the millimeter range. The hysteresis curves of both materials show the same shape and are indistinguishable in spite of different RRRs ($RRR_{Wah\ Chang}$ = 700, $RRR_{Heraeus}$ = 380). The curves are reversible above a magnetic field of 140 mT and have a negligible zero-field magnetization of about 5 mT. Hence, a high degree of homogenization together with a small amount of flux pinning is reached after the 1400 °C heat treatment. This is independent of the method used to remove the damage layer (BCP of 70 μm or 170 μm, electropolishing or grinding). Also no difference was seen whether the annealing at 800 °C had taken place or not.

The removal of the titanium layer, which has a thickness of about 10 μm and a penetration depth by diffusion in the grain boundaries of up to 60 μm, has no influence on the results of the magnetization measurements. Hence, this method is predominantly sensitive to the bulk properties.

The rather small amount of flux pinning after the 1400 °C heat treatment enables a reasonably accurate determination of the lower critical field B_{c1}. From Fig. 4 the magnetic field at maximum magnetization is about 110 mT. The demagnetization factor of the samples is estimated to be 1.2-1.3, yielding a lower critical magnetic field of 130-140 mT at 4.2 K.

B. Susceptibility Measurements

The susceptibility curves of the Wah Chang and the Heraeus niobium samples after the different consecutive preparations are shown in Fig. 7. The untreated samples show two transitions from the superconducting to the normal conducting state. The first transition starts at ≈ 270 mT and is correlated with the upper critical magnetic field in the bulk as derived from the magnetization measurements above. The second transition extends from 550 mT to about 800 mT. This enhanced critical magnetic field is probably associated with the surface damage layer which contains a large amount of impurities and lattice defects.

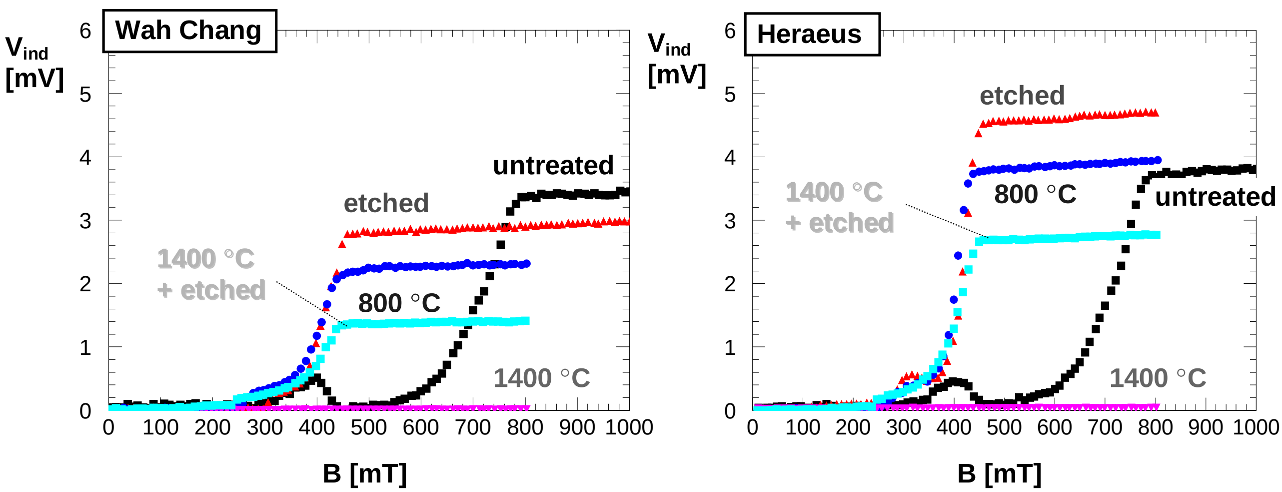

Figure 7: Susceptibility measurements of the Wah Chang (left) and Heraeus (right) material untreated and after different consecutive preparation steps (BCP, annealing at 800 °C, heat treatment at 1400 °C plus titanium getter, BCP).

After removal of the damage layer by chemical etching the transition width from the superconducting to the normal state is strongly reduced extending now from 270 to 450 mT. The fact that one does not observe a steep step to normal conductivity at the critical field B_{c2} ≈ 270 mT of the bulk but a widened transition region between 270 and 450 mT may be an indication of the appearance of the surface critical field B_{c3} whose value is 1.69*B_{c2} according to the Ginzburg-Landau theory.

The 800 °C annealing leaves the transition width invariant. After the 1400 °C high temperature treatment with titanium getter no step to normal conduction is observed up to a dc field of 2 T. This behavior is caused by a Nb_xTi_y surface layer which may have upper critical fields in the many tesla range. If the titanium layer is removed the transition in the 270-450 mT range is recovered.

The step height of the transition is related to the RRR. Calculations considering the skin effect in normal conducting niobium result in a behavior proportional to exp (-d*RRR$^{1/2}$), where d is the sample thickness, as it is shown in Fig. 8.

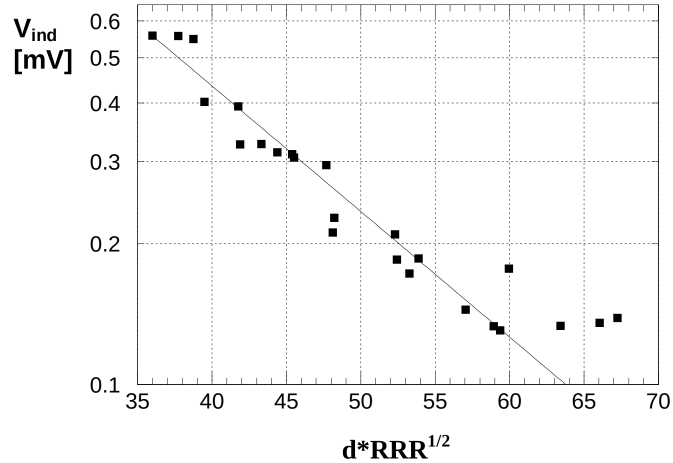

Figure 8: Step height observed in the susceptibility measurements of the transition from the superconducting to the normal conducting phase in dependence from the sample thickness *d* and the RRR value.

IV. Conclusion

Niobium samples have been investigated to check all steps of cavity preparation for the TTF project. Magnetization and susceptibility measurements were used to examine the critical magnetic fields and the magnetic flux pinning, especially the dependence on chemical etching and heat treatment. By the first BCP the damage layer is removed leading to a reduction of flux pinning at the sample surface. The first homogenization of niobium of different origin could be noticed after the annealing at 800 °C, leading to comparable grain sizes and number of grain boundaries in all samples. Also internal stress by plastic deformation has been removed. The high temperature heat treatment at 1400 °C together with titanium getter plus subsequent BCP led to the outdiffusion and removal of impurities and thus to an improvement of the RRR. Also a considerable growth of the grain size has been observed. Because of this reduction of pinning centers the niobium has only a small amount of flux pinning, the magnetization behavior is almost reversible. Combined with the fact that also the RRR is significantly increased this may be the explanation for the often observed improvement in cavity performance after a 1400 °C heat treatment.

V. Acknowledgement

The authors would like to thank O. Peters and his group for building the test setup, H. Brück and M. Stolper for the help in setting up the electronics and the data analysis system.

VI. References

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