RESEARCH ON SUPERCONDUCTING NIOBIUM CAVITIES AT BROOKHAVEN*

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Abstract

Recent experiments on superconducting niobium cavities were aimed primarily at achieving high Q's at high rf field levels and at producing more stable surfaces. Best results were obtained with electron-beam-welded cavities machined from large grain billets and outgassed above 1800°C in the furnace at the High Energy Physics Laboratory at Stanford. The highest surface magnetic and electric fields were $\hat{B} \approx 550$ G and $\hat{E} \approx 21.5$ MV/m measured in the TM_{010} and TM_{110} modes, respectively. Improvement factors in all modes between 2.3 and 4.1 GHz were above 10^5. Anodized cavities exhibit initially higher Q's but they deteriorate when kept at room temperature even in a good vacuum. Anodic films of Nb_2O_5 are therefore not suitable as protective surfaces at room temperature. In order to gain some information on the influence of irradiation on Q's and $\hat{B}$'s, the cavities were placed in the direct beam of Brookhaven's 200-MeV proton linac. Unoxidized cavities show a decrease of about 40% in residual Q's when irradiated with $\sim 10^{15}$ protons/cm^2. Much more severe deterioration in both Q's and peak fields occurs in anodized cavities. These measurements represent the only radiation damage data on superconducting microwave devices.

1. Introduction

In the near future the most important applications of superconductivity at microwave frequencies will be RF beam separators and linear accelerators. The success of these machines will ultimately depend on high Q iris-loaded cavities with high accelerating and deflecting gradients. After extensive work on lead-plated copper and solid niobium cavities in many laboratories, niobium emerged as a leading candidate for construction of iris-loaded structures. These structures are conveniently manufactured either by electroforming or by electron-beam (EB) welding of small sections which are machined from large crystal billets or from hydroformed cups.

Since the RF fields interact with the material in a thin layer (~ 500 Å) only, the finished cavities are subjected to several treatments resulting in smooth and clean surfaces[1,2] in order to achieve high Q's and high levels of electromagnetic fields on cavity walls. Smooth and clean surfaces can also be reacted with other elements, namely Sn and Al (Ref. 3) to produce thin films of niobium compounds with high T_c. Thus far only oxide films were investigated[4-6].

Both Q's and peak fields are susceptible to a deterioration when exposed to the atmosphere or when they are operated at poor vacuum. An additional degradation takes place when the cavities are irradiated[7]. The above topics will now be covered in some detail.

2. Comparison of Experimental Results

The most important properties a cavity should possess in order to be useful for linear accelerators or RF separators are:

1) High residual Q (Q_{res}) since the operating temperature of practical devices is $T \approx 1.8$°K.

2) Capability of maintaining high RF fields on its walls.

3) Negligible degradation of the above values with time.

Q_{res} and RF fields were measured by transmission method[8] in cavities whose geometry is shown in Fig. 1. A sufficient number of modes was investigated to study effects of high RF fields in the region of EB welds. The highest measured RF magnetic field perpendicular and parallel to EB welds were $\hat{B}_\perp \approx 548$ G and $B_\parallel \approx 376$ G. In the case of $B_\parallel$ much higher fields of 554 G existed elsewhere in the

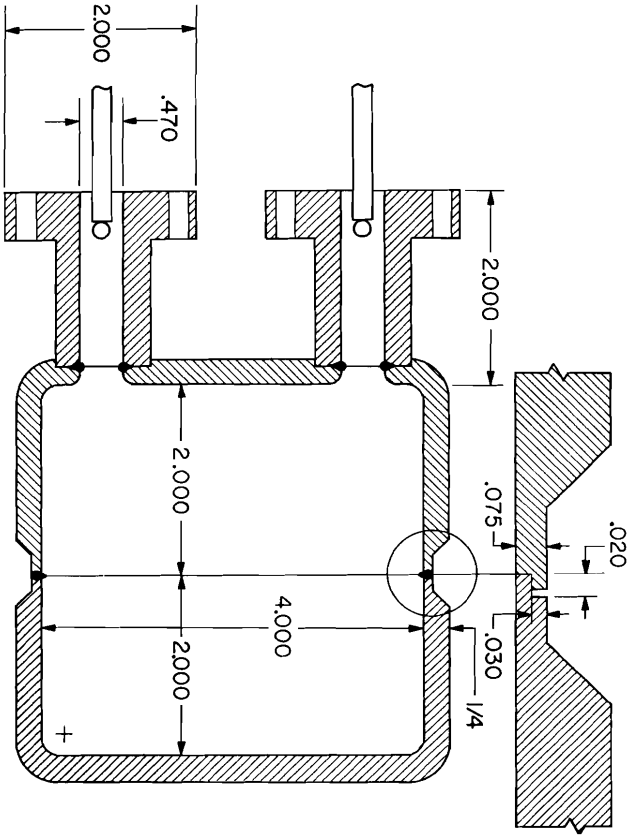

Fig. 1. Niobium cavity with its coupling loops. Dimensions are in inches; dots represent EB welds.

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cavity. The highest value of RF electric field normal to the weld was $\hat{E} \approx 11.7$ MV/m, at which point magnetic breakdown occurred. In all cases, the input power to the cavities was limited by an abrupt transition from superconducting to normal state caused by the formation of normal nucleii due to local RF heating and low specific heat of superconducting Nb. At 2°K $C_p = 1.5 \times 10^{-5}$ J/g-°K (Ref. 9). With the exception of a single case of deteriorated Nb_2O_5 film, to be discussed in the next Section, X-radiation was never observed at the highest field levels. We, therefore, conclude that this limit is of a magnetic nature, and we call it the breakdown RF magnetic field $\hat{B}_B$.

The best results obtained in cavities manufactured by three techniques suitable for practical devices are compared in Table I. All cavities were outgassed at least once in a high-vacuum, high-temperature furnace at HEPL or SLAC, from where they were shipped sealed with indium gaskets under nitrogen atmosphere. Upon arrival at Brookhaven they were opened and exposed to some room atmosphere due to the difficulties with the removal of indium gaskets from the flanges. This accounts for lower Q's (Table I) as compared with the S-band cavities at SLAC, where $Q \approx 1.3 \times 10^{10}$ and $\hat{B} \approx 390$ G were measured at 1.85°K (Ref. 10).

It is apparent from Table I that EB-welded Nb cavities machined from large grain material are superior and yield the highest $\hat{B}_B = 554$ G in the TM_{010} mode, while the highest $\hat{E}$ of 21.5 MV/m is achieved in the TM_{110} mode. In the TM_{010} and TM_{011} modes a field enhancement of 40% due to the coupling holes is used[11].

3. Anodic Oxidation

Following the announcement of improvement in the behavior of anodized Nb cavities[4,5] a similar investigation was undertaken at BNL[6]. Niobium cavities (Fig. 1) were oxidized in two different solutions, namely 0.2 NH_2SO_4 (Ref. 12) and ~ 20% solution of ammonia[4]. Oxide films of 200-1000 Å were formed both at slow and fast rates. The best measured Q_{res} and $\hat{B}_B$ are compared with other surfaces in Table I. It is seen that anodized cavities exhibit initially higher Q's which might be due to the conversion of metallic NbO as well as other Nb suboxides and dissolved oxygen into Nb_2O_5, a low-loss insulator[6]. Lower Nb oxides including NbO form inadvertently on Nb surfaces when the cavities are cooled even in an ultra-high-vacuum furnace[13]. Unfortunately these high Q's deteriorate when the cavities are kept at room temperature even in good vacuum of 10^{-7} Torr. This behavior of S-band cavities differs substantially from X-band cavities measured at Siemens[14] where no aging has been observed. Even though the mechanism of aging is not well understood there seems to exist a strong evidence[6] that Nb_2O_5 decomposes into lower oxides plus oxygen which diffuses into Nb, producing a lower T_c surface layer[15]. Both reactions, coupled with changes in the oxide itself, tend to decrease residual Q's and peak fields. Several aging experiments for various oxide films are depicted in Fig. 2. From these as well as from other tests not reported here, thinner films ~ 200 Å formed more rapidly seem to exhibit least deterioration with time.

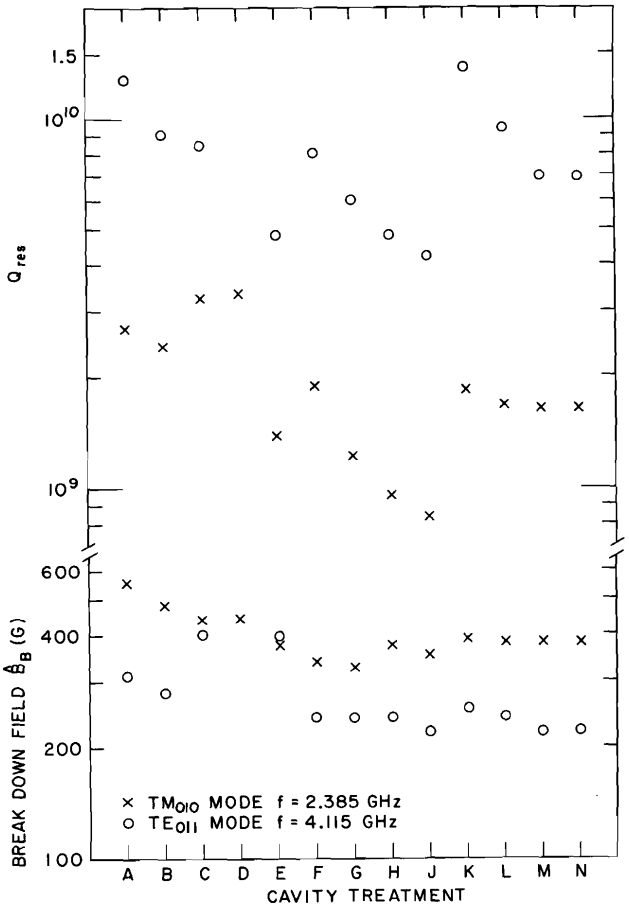

Fig. 2. Changes of Q_{res} and $\hat{B}_B$ due to aging of various Nb_2O_5 films in a large grain Nb cavity. A: outgassed at 1850°C in 5×10^{-8} Torr. B: anodized in 0.2 NH_2SO_4 to 360 Å (2 min). C: chemically polished and anodized in 0.2 NH_2SO_4, 360 Å (2 min). E: five days at 20°C and 9×10^{-7} Torr plus 19 h at 20°C in air. F: chemically polished and anodized in 20% solution of NH_3, 950 Å (9 min). G: processing 45 min. H: 36 h at 20°C, 1×10^{-7} Torr. J: processing (operating a cavity continuously at power levels where breakdowns occur is called processing) 100 min. K: chemically polished and anodized in 0.2 NH_2SO_4, 200 Å (1.5 min). L: processing 50 min. M: five days at 20°C, 3×10^{-7} Torr. N: processing 65 min.

In addition to aging, one cavity oxidized in 0.2 NH_2SO_4 for 19 s with resulting thickness of 200 Å developed X-radiation (Fig. 3) after two test runs and 15 days at 20°C in 10^{-8} Torr range. Significant radiation (25 mR/h) outside the dewar was observed in the TM_{010} mode only (Fig. 3). The resonant frequency of the cavity decreased by 25 Hz by going from a low field $\hat{E} = 4$ MV/m to $\hat{E} = 13.5$ MV/m. A further increase of input power resulted in the characteristic magnetic breakdown. Much higher electric field ≈ 20 MV/m was reached in TM_{110} (f = 3.8 GHz) with negligible radiation. According to Halbritter[16] the X-rays and the accompagnying break-down can be attributed to second order multipactoring.

TABLE I. Comparison of Experimental Results of Niobium Cavities

Manufacturing Process		^{o}K	TM_{010}	TE_{111}	TM_{011}	TM_{110}	TE_{011}	
Frequency			2.25	2.3	2.7	3.6	3.9	GHz
Electroforming	Best Q_{res}	1.4	1.5	3.4	0.8	1.6	27.4	10^9
	Highest $\hat{B}_B$	1.4	142	290	142	141	327	G
	Highest $\hat{E}$	1.4	5.2	6.2	4.4	8.5		MV/m
Hydroformed cups machined and EB welded	Best Q_{res}	1.4	4.2	8.4	1.9	2.4	13.1	10^9
	Highest $\hat{B}_B$	1.4	464	459	357	313	345	G
	Highest $\hat{E}$	1.4	17.1	9.8	11.0	18.8		MV/m
Frequency			2.39	2.42	2.9	3.8	4.12	GHz
Large crystal billets machined and EB welded	Best Q_{res}	1.4	4.8	10.4	2.7	8.7	26.7	10^9
	Highest $\hat{B}_B$	1.4	554	548	362	358	323	G
	Highest $\hat{E}$	1.4	20.4	11.7	11.1	21.5		MV/m
EB-welded cavities oxidized in 0.2 NH_2SO_4	Best Q_{res}	1.4	5.1	12.1	3.9	5.9	29.0	10^9
	Highest $\hat{B}_B$	1.4	485	491	325	313	400	G
	Highest $\hat{E}$	1.4	17.8	10.5	10.0	18.8		MV/m

TABLE II. Effects of Radiation Damage on Anodized Superconducting Niobium Cavities

		^{o}K	TM_{010}	TE_{111}	TM_{011}	TM_{110}	TE_{011}	
Frequency			2.258	2.297	2.726	3.603	3.891	GHz
Geometric constant	QR		302	321	271	481	780	Ω
Before irradiation	R_t	4.2	1.22	1.28	1.78	3.1	3.8	μΩ
After irradiation	R_t	4.2	1.2	1.28	1.97	3.07	3.8	μΩ
After chemical repolishing	R_t	4.2	1.22	1.29	1.77	3.07	3.9	μΩ
Before irradiation	Q_{res}	1.4	2.95	12.1	3.17	2.85	17.2	10^9
	$\hat{B}_B$	1.4	314	353	293	262	197	G
After irradiation	Q_{res}	1.4	0.29	0.31	0.16	0.25	0.7	10^9
	$\hat{B}_B$	1.4	243	236	246	202	150	G
After annealing for 80 h at room temperature	Q_{res}	1.4	0.24	0.34	0.15	0.21	0.74	10^9
	$\hat{B}_B$	1.4	215	218	258	206	172	G
After chemical repolishing	Q_{res}	1.4	1.19	3.1	0.83	1.0	3.8	10^9
	$\hat{B}_B$	1.4	296	320	274	221	180	G

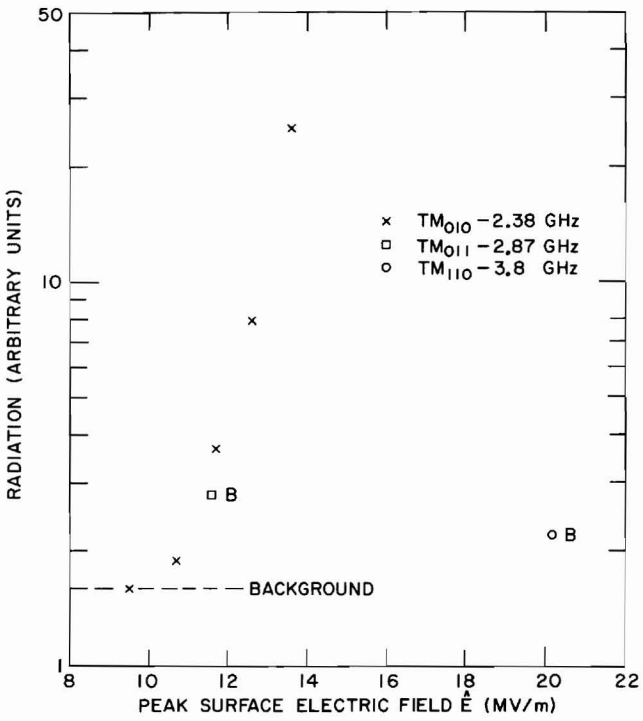

Fig. 3. X-radiation in mR/h outside the dewar vs peak surface electric field in an oxidized cavity after 15 days of aging. The background in the dewar was caused by previous irradiation.

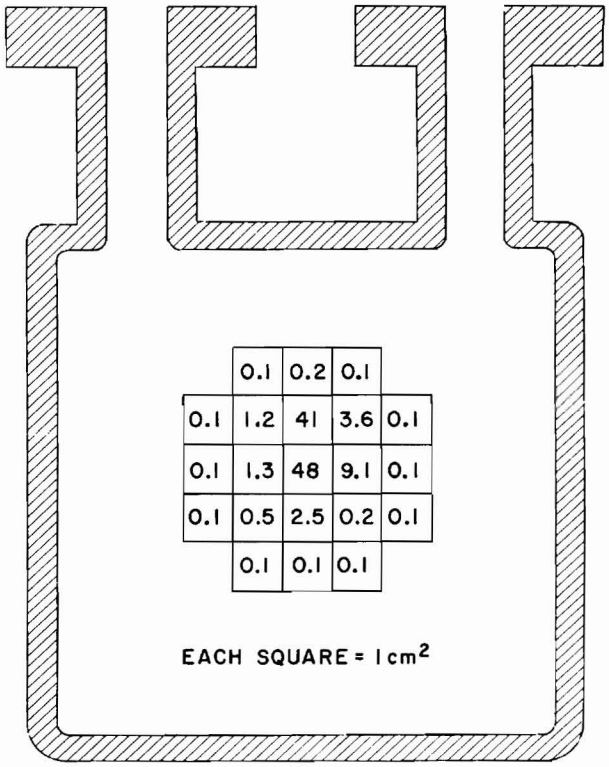

Fig. 4. Distribution of integrated beam flux in 10^{14} protons/cm^2.

4. Radiation Damage

Effects of radiation on Q's and $\hat{B}_B$'s were studied in hydroformed EB-welded cavities both with and without oxide films. Unoxidized cavities[7] showed about a 40% decrease in Q_{res} when irradiated with 173-MeV protons from the 200-MeV linac. Total flux of 10^{16} protons was used with a density distribution of 10^{15} p/cm^2.

After these experiments[7] the radioactive cavity was stored at atmospheric pressure. Seventy days later it was chemically repolished ($\sim$ 20 μm of surface was removed) and anodized in 0.2 NH_2SO_4. 320 Å film was formed in $\sim$ 18 s. Measured residual Q's at 1.4°K were substantially higher than those reported in Ref. 7 due to Nb_2O_5 film, but $\hat{B}_B$'s were essentially unchanged. The cavity with its dewar and measuring apparatus was then placed into a direct beam of the BNL 200-MeV linac and irradiated with $\sim 10^{16}$ protons. Before the beam reaches the cavity it must pass through the stainless-steel dewar, μ-metal shield, liquid nitrogen and liquid helium, which degrade both its energy and intensity[17]. The distribution of the remaining 180-MeV protons incident on the cavity (Fig. 4) was obtained by exposing 0.003-in. Al foil. The absolute number of protons measured by linac current transformer was found to be in good agreement with the number obtained from the exposed Al foil. During the whole time between the measurements before and after irradiation the cavity was submerged in liquid helium, to minimize any additional changes in Q's due to possible chemical reactions of condensed gases with Nb surfaces upon warm-up.

Inspection of Table II reveals a drastic decrease in all Q_{res}'s due to irradiation with no adverse effects on the superconducting part of surface resistance R_t. After these measurements the cavity was warmed up to room temperature and kept there for $\sim$ 80 h while the pressure stayed between $1 - 3 \times 10^{-7}$ Torr. As seen from Table II no noticeable improvement resulted from this annealing, since the radiation damage was caused primarily by the deterioration of the oxide film. Even the removal of 20 μm by chemical polishing followed by anodizing failed to restore the high residual Q's measured previously. From the above experiments we can conclude that the interacting high energy protons tend to break up the Nb_2O_5 films which results in oxygen diffusion into niobium and formation of lower oxides in a similar way as discussed under aging in the previous Section. For the case of radiation, however, the oxygen diffuses much faster and deeper, with much more severe effects.

5. Conclusions

1) The best cavities of today are those machined from large grain Nb, outgassed above 1850°C in 10^{-8} Torr and sealed off if possible. Such cavities not only attain the highest Q's and peak fields but also show negligible aging.

2) High electric and magnetic RF fields can be maintained in the region of EB welds without excessive dissipation or field emission. From Section 2 $\hat{B}_{\perp} \approx 550$ G, $\hat{B}_{\parallel} > 367$ G and $\hat{E} > 11.7$ MV/m.

3) Aging seems to be caused primarily by chemical reaction of various materials present on the surface, notably oxygen, with Nb surfaces. Nb_2O_5 which makes oxygen available at the metal-oxide interface is therefore not a suitable protective film for Nb cavities kept at room temperature for long periods of time. Additional objections to Nb_2O_5 are greater radiation damage and enhanced electron emission. Other protective films have not yet been investigated in RF resonators but M. Dickey and M. Strongin succeeded in producing good Nb_3Sn and Nb_3Al films on flat Nb samples. Nitride films, on the other hand, were unacceptable. In the search for more stable surfaces we plan to react our cavities with Sn and Al in cooperation with M. Strongin, M. Dickey and H. Farrell to produce thin surface films of Nb_3Sn and Nb_3Al later this year when the UHV furnace is installed.

4) Radiation, like aging, produces the greatest degradation in Q's and peak fields in cavities whose surfaces are covered by various oxides and other contaminants or contain large amounts of interstitial oxygen. If we could produce perfectly clean surfaces, radiation damage, at least for exposures up to a few times 10^{15} protons/cm^2, would be negligible.

6. Acknowledgements

I would like to acknowledge discussions on niobium surfaces and thin films with Drs. M. Strongin, M. Dickey and H. Farrell, as well as discussions on anodic oxidation with Dr. H. Isaacs. The technical assistance of Mr. F. Timm in all phases of the project was greatly appreciated. The microwave measurements were carried out in cooperation with Mr. S. Giordano. Furthermore, I would like to thank Dr. W. Moore for many enlightening discussions on radiation damage. Finally, I am very grateful to Drs. Fred Mills and John Blewett for their generous support of this research.

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DISCUSSION

H. DIEPERS : It is known from the literature that one gets a very homogeneous Nb_2O_5 layer, if one anodizes with a low current density. What was the current density in your experiment ?

H. HALAMA : I tried various current densities from very low up to ~ 4mA/cm^2. The most serious ageing (degradation) was observed in a film 950 Å thick formed in a 20% solution of NH_3 for 9 minutes. The starting current was 1mA/cm^2. This film also shows a Q degradation during processing.

P.R. TUNNICLIFFE : Was your anodized cavity held at liquid helium temperatures when you observed Q degradation with time ?

H.HALAMA : Residual Q degradation in cavities held at liquid He and liquid N temperatures was negligible.

H.A. SCHWETTMAN : Can you quote tolerable radiation levels for niobium cavities that are UHV fired and for niobium cavities that are anodized.

H. HALAMA : Radiation produces greatest degradation in cavities whose surfaces are covered with oxides or other contaminants. It is difficult to quote the tolerable levels based on three existing experiments but I believe that the usefulness of a clean cavity heat-treated in your furnace would not be appreciably affected by exposures up to 10^{16} protons/cm^2. The

anodized cavity, on the other hand, would not tolerate 10^{15} protons/cm^2.

A. CITRON : Speaking of degradation of Q values, there are several mechanisms that reduce initially very high Q values to more modest ones. But the question is whether the low values are still practically acceptable. In the case of the ageing you observed, it might well be that the Q levels off at acceptable values.

H.HALAMA : I have not studied the ageing over long enough periods of time (many months) to answer your question properly. In some cavities the degradation over 15 days was not too serious; i.e., the cavities would still be good for practical applications.