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Expected dependence of Nb-coated RF cavity performance on the characteristics of niobium

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Abstract

Compared to traditional superconducting cavities made of niobium sheets, Nb-coated copper cavities present higher Qo values at low fields but unfortunately also a faster Qo degradation with increasing field.
This degradation is very detrimental to cavity performance because it results in higher cryogenic losses and may even limit the operating field in the absence of other limiting effects such as quenching or electron field emission.
The discussion reported in this paper suggests that Qo degradation may originate at the Nb film grain boundaries, and that a substantial improvement could be obtained by increasing the Nb grain size.
It is also shown that Nb grains of the desired size have been obtained by a realistic increase in the coating temperature and the Nb film thickness.

1. *Introduction*

In the framework of a development programme stimulated by the needs of the CERN LEP 200 project, the feasibility of Nb-coated copper accelerating cavities has been studied and established [1, 2].

With respect to cavities traditionally made of niobium sheets, coated cavities provide a higher stability against quench, higher unloaded quality factor Qo values in the operating field range, and insensitivity to earth magnetic field trapped while cooling. Because of these advantages, it has been decided to order about 170 Nb-coated cavities for upgrading LEP beams energy from 55 to about 90 GeV. These cavities, presently produced by three European manufacturers, should be installed in LEP before the end of 1995 [3].
Unfortunately however the Nb-coated cavities also present an important degradation of Qo consequent to increasing the accelerating field E; this degradation results in Qo values of about 4 to 5 x 10^9 at 6 MV/m, while at low field values up to 10^{10} have been measured.

Since the power dissipation in a cavity varies as E^2/Qo, the cryogenic cooling power installed for LEP would permit operating Nb-coated cavities up to 10 MV/m if their initial Qo could be preserved. Therefore a large effort has been devoted to study the Nb-coated cavity behaviour in order to possibly reduce Qo deterioration. The status of our present understanding of the physics underlying this problem is summarized below.

2. *RF superconducting surface resistance at low field*

In contrast to displaying zero resistance when a d.c. voltage is applied, superconductors present a finite (surface) resistance when exposed to RF power due to "normal conducting" electrons, always present at temperatures above absolute zero.

For any given frequency and temperature the BCS theory of superconductivity allows the surface resistance R_S of a superconductor to be calculated as a function of the electron mean free path ℓ. This calculation shows that R_S reaches a minimum for $\xi_0/\ell \approx 1$, where ξ_0 is the coherence length [4].

The result of this calculation for Nb at 4.2 K and 352 MHz frequency is shown in Fig. 1, where R_S has been replaced by Qo and ℓ by the residual resistivity ratio RRR (defined as the ratio of the resistivity measured at room temperature to that measured just above transition temperature, and therefore proportional to ℓ). This change of variables facilitates the interpretation of the plot because Qo and RRR are the quantities directly measured.

Also shown in Fig. 1 is the BCS contribution to the Qo values measured for Nb sheet and Nb-coated LEP cavities. The comparison indicates that calculations provide the right trend but not the right absolute values of Qo. A similar conclusion was already reached for lead coated cavities at 3 GHz [4] and for Nb cavities at 500 MHz [5]. The reason for this discrepancy may be found in the strong coupling behaviour of Nb which is not properly described by the BCS standard formalism, which predicts $2\Delta/kT_C = 3.56$. The curve reproduced in Fig. 1 has instead been calculated by replacing the BCS value with a strong coupling factor of 3.7. In the limit of validity of this approximation, a good fit to the experimental data is obtained by using a strong coupling factor of 4.3. Figure 1 also shows that a better agreement with the experimental data may be obtained by using the two fluids model in the dirty limit approximation [6], which however is only valid for $\ell < \xi_0$.

In spite of the theoretical difficulties, the above discussion provides convincing evidence that the higher Qo value of the Nb-coated cavities is a direct consequence of the Nb film RRR value being lower than that of bulk Nb. By chance the presently achieved RRR value of about 15 provides the highest possible Qo. However, the broad maximum of the Qo (RRR) curve would allow increasing RRR considerably without appreciably reducing Qo.

3. *Qo degradation*

Various models have been produced to account for the Qo degradation observed in Nb-coated cavities with increasing accelerating field [7, 8, 9]. They were derived from other models developed for HTc films [10, 11] under the compelling evidence of an RF field amplitude dependence of granular film surface resistance and of the lack of such dependence for single crystals and/or epitaxially grown films [12, 13].

In spite of some differences, which however cannot be discriminated experimentally and therefore are not relevant in the context of this discussion, the proposed models describe the superconducting film as a network of superconducting grains coupled by Josephson junctions at grain boundaries.

This implies the presence of a temperature independent (i.e. non BCS) component of the surface resistance (or residual resistance R_{res}) which increases with the applied RF field amplitude.

The practical relevance of this model consists in stipulating that Nb films characterised by a lower grain boundary density (i.e. larger grain size) should provide better RF performance with respect to Qo degradation.

As shown in paragraph 4, grain boundaries also provide a substantial contribution to the low temperature resistivity in the normal conducting state. Consequently, the guide-line to produce better cavities consists in increasing the RRR of the Nb film. As an additional and independent proof of this conclusion, it is worth recalling that a smaller Qo degradation has been noticed in the past each time an RRR increase has been achieved [2].

The expected performance of a Nb-coated LEP cavity at 4.2 K is here anticipated in Fig. 2. In this figure the Qo (E) curve measured on a cavity of average quality has been corrected by calculation for an RRR increase from 15 to 25 and 35. The procedure followed to correlate RRR values to grain size, which defines the RF performance, is described in paragraph 4.

Figure 2 indicates that the RRR = 25 curve presents a large reduction of slope without a noticeable decrease of the initial Qo value, while further increasing RRR is less beneficial on the slope and reduces more appreciably Qo at low (zero) field. Consequently, it may be concluded that RRR values in the range from 25 to 35 provide the best compromise for the application, as well for any other application where R_{res} is much smaller than the BCS R_S. When this condition does not apply, as it is the case for cavities operating at about 2 K, higher RRR values may be required because in this case R_S is negligible and the cavity performance depends mainly on the residual resistance.

4. *The RRR of polycrystalline thin films*

The mean free path ℓ of electrons in a metal may be limited by phonons, crystal defects (impurities, dislocations, etc.) and grain boundaries.

At very low temperatures the phonon density becomes negligible and ℓ is limited either by defects or grain boundaries depending on the relative magnitude of the limit set by defects (ℓ_∞) and on average crystal grain size D. It has been shown [14] that the low temperature resistivity of a polycrystalline film may be written as :

$$\rho = \frac{m v_F}{n e^2 \ell_\infty} T^{-\ell_\infty / D} \qquad (1)$$

where T is the probability for an electron to pass a single grain boundary. This simplified formulation provides a good fit to experimental data for $T > 0.6$, i.e. for relatively clean grain boundaries, easily crossed by electrons. Since T is always smaller than 1, eq. (1) indicates that larger grains result in lower resistivity; for increasing D, ρ approaches asymptotically the intragrain resistivity.

The mechanism of grain growth has been studied by Monte Carlo simulation on the ground of thermodynamic and kinetic considerations [15, 16].

The results of computation indicate that at a given temperature the grain size D increases with increasing film thickness z according to the following exponential law:

$$D = a z^b \qquad (2)$$

where b is approximately 0.4 [16].

The computer simulated film cross section displays a columnar structure along which the smaller grains tend to disappear in the larger grains.

When combining the described resistivity dependence on grain size and the dependence of the latter on film thickness, a comprehensive description of the RRR dependence on film parameters may be obtained.

By fitting the resulting formulation to experimental data, important physical information may be obtained, namely average grain size, intragrain resistivity and probability of electron transmission at grain boundaries. This analysis shows that in reality the RRR value of a thin film is not uniform, but increases when moving from the substrate to the surface until it is stabilised by defects and impurities. Since the intensity of the RF field decreases exponentially below film surface, the effective RRR of the film as experienced by the RF field is larger than the measured one.

A correct estimate of a Nb-coated cavity performance should take the RRR variation into account by means of an appropriate weight factor. This correction has been indeed applied for the estimates reported in Fig. 2.

5. *Experimental*

On the ground of the arguments reported above, the experimental activity has been recently focused on studying the dependence of RRR on the coating conditions, such as coating procedure, film thickness, discharge parameters and coating temperature.

The study has been carried out on samples (quartz and copper) coated inside stainless steel cavities (single-cell 500 MHz or four-cell 352 MHz) equipped with sample holders [2].

The final goal of the study consists in defining one or two coating processes providing reproducibly RRR values higher than about 15 (present value obtained inside LEP cavities of industrial production) which could eventually be implemented on cavity coating for RF performance evaluation and later on possibly applied to the industrial production.

The results obtained so far are the following.

5.1. *Coating procedure*

According to the standardized coating procedure [2], the tubes at both extremities of a cavity are coated first and subsequently cavity cells are coated in the order 1 - 4 - 2 - 3 (in this numbering 1 indicates the top cell and 4 the bottom cell in the vertical coating mounting).

It has been observed, initially on samples produced at CERN and later by all cavity manufacturers [2], that the RRR values obtained in cells 1 and 4 were systematically lower than those of cells 2 and 3 (about 13 instead of about 16).

This difference has been eliminated and RRR has been increased uniformly up to about 20 by sputter cleaning the cathode over its whole length before carrying out sample coating.

The cause of this behaviour was found in the Nb film contamination produced by degassing of the cathode surfaces between magnets, which are not properly cleaned during the standard coating process [2].

Single-cell 500 MHz cavities are less sensitive to this effect due to the shorter cathode and smaller distance between magnets.

5.2. *Film thickness*

The RRR dependence on Nb film thickness has been studied on a single-cell 500 MHz cavity equipped with two sample holders placed at diametrically opposite points of the cavity equatorial plane. Each sample holder could in turn expose six different samples for coating by means of a carrousel device.

The results obtained are shown in Fig. 3. The average grain size measured by electron microscopy on the sample surface is 0.1 μm for a 2 μm thick sample. By fitting eq. (2) to the measured grain sizes for films of different thickness (expressed in μm), one obtains $a \approx 0.07$ and $b \approx 0.34$. Using these values in eq. (1) gives $T = 0.67$, intragrain RRR ≈ 55. Note that the quoted intragrain resistivity corresponds [17] to the value obtainable in bulk Nb loaded with H_2 at a concentration of about 0.4%, a value very close to that measured on one of our samples [18].

Figure 3 indicates that the thickness of about 1.3 μm standardised for cavity coatings, although redundant to cope with RF penetration in superconducting Nb (for which the penetration depth is about 40 nm), should not be reduced but rather increased to provide higher RRR values. For instance an RRR value of 25 could be obtained by simply increasing the film thickness up to about 3 μm.

5.3. *Discharge parameters and coating temperature*

The sputtering discharge characteristics depend on cathode voltage, magnetic field (i.e. solenoid current), argon pressure. The variation domain of these parameters was explored and the most relevant results are presented in Fig. 4 where the RRR values obtained are plotted as a function of the argon pressure for the initially standardised situation (400 V, discharge power 6.3 kW) and for the best situation so far obtained (400 V, 11 kW). It should be noted that in the latter case the growing rate of the Nb film is about two times higher due to the larger discharge power. The results of Fig. 4 clearly indicate that sample RRR's are very sensitive to a cavity coating temperature increase from 200 °C to 300 °C. Direct testing and stability computations by finite elements method show that a temperature of 300 °C is tolerable during the short duration of the sputtering process (less than 1 hour per cell).

The results displayed in Fig. 4 are relative to samples placed on sample holders installed in cells 2 and 3 and produced without cathode sputter cleaning which, when applied, results in an additional increase of RRR (see paragraph 5.1).

Even larger RRR values were obtained on samples lying on the flat lower part of cavity cells. In this case, for a cavity temperature of 300 °C and the best coating conditions indicated in Fig. 4, RRR values ranging from 35 to 40 were measured. These very high values were obtained for films about 4 μm thick and it was found that they were caused by the high temperatures (550 °C) reached by the sample during coating due to its poor thermal contact with the cavity wall. The grain size at the surface of these samples is about 0.2 μm with isolated randomly distributed grains of larger size (about 0.5 μm).

6. Conclusions

Nb-coated cavities are today an industrial reality. In the version developed for LEP 200 they provide better performance at lower cost than niobium sheet cavities. Unfortunately, however, they suffer from a faster Qo degradation when increasing the accelerating field.

A considerable effort has been devoted to understand the physics underlying the behaviour of the Nb-coated cavities so as to possibly reduce this inconvenience. To the best of present knowledge, Qo degradation seems to originate at Nb crystal grain boundaries.

Consequently, it should be possible to reduce it by growing films with a lower concentration of (cleaner) grain boundaries. A relevant conclusion of the study described in this report is that both cavity performance and film quality may be related to the same physical quantity, the low temperature resistivity of Nb in the normal state or its equivalent expression RRR. A systematic optimization of all aspects of the preparation of cavities indicates that Nb coatings characterised by RRR values in the range from 25 to 30 should be feasible. This should considerably increase cavity Qo at accelerating fields higher than about 3 MV/m. The validity of this conclusion will be checked soon by means of some cavities which will be coated at CERN according to the optimised procedure. These cavities will also indicate if a Nb film of better quality will remain insensitive to trapped magnetic field and if larger grains with sharp edges will not be detrimental with respect to electron field emission.

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Figure Captions

Fig. 1 Variation of the BCS Qo for Nb at 4.2 K as a function of RRR or electron mean free path. The triangles represent the values estimated according to the BCS theory but making use of a strong coupling factor of 3.7. Circles represent the estimation carried out in the dirty limit approximation. The experimental values are given with error bars representing the maximum spread of values measured on 4 (Nb sheet) or 12 (Nb-coated) cavities.

Fig. 2 Estimated improvement of cavity performance consequent to Nb film RRR increase from 15 (average value for the present industrial production) to 25 or 35. For these estimates also the Qo variation shown in Fig. 1 is taken into account.

Fig. 3 Variation of RRR as a function of the Nb film thickness. The dotted line represents the best fit to the experimental values computed as discussed in paragraph 4.

Fig. 4 Variation of Nb film RRR obtained for various discharge parameters and cavity temperatures during coating.

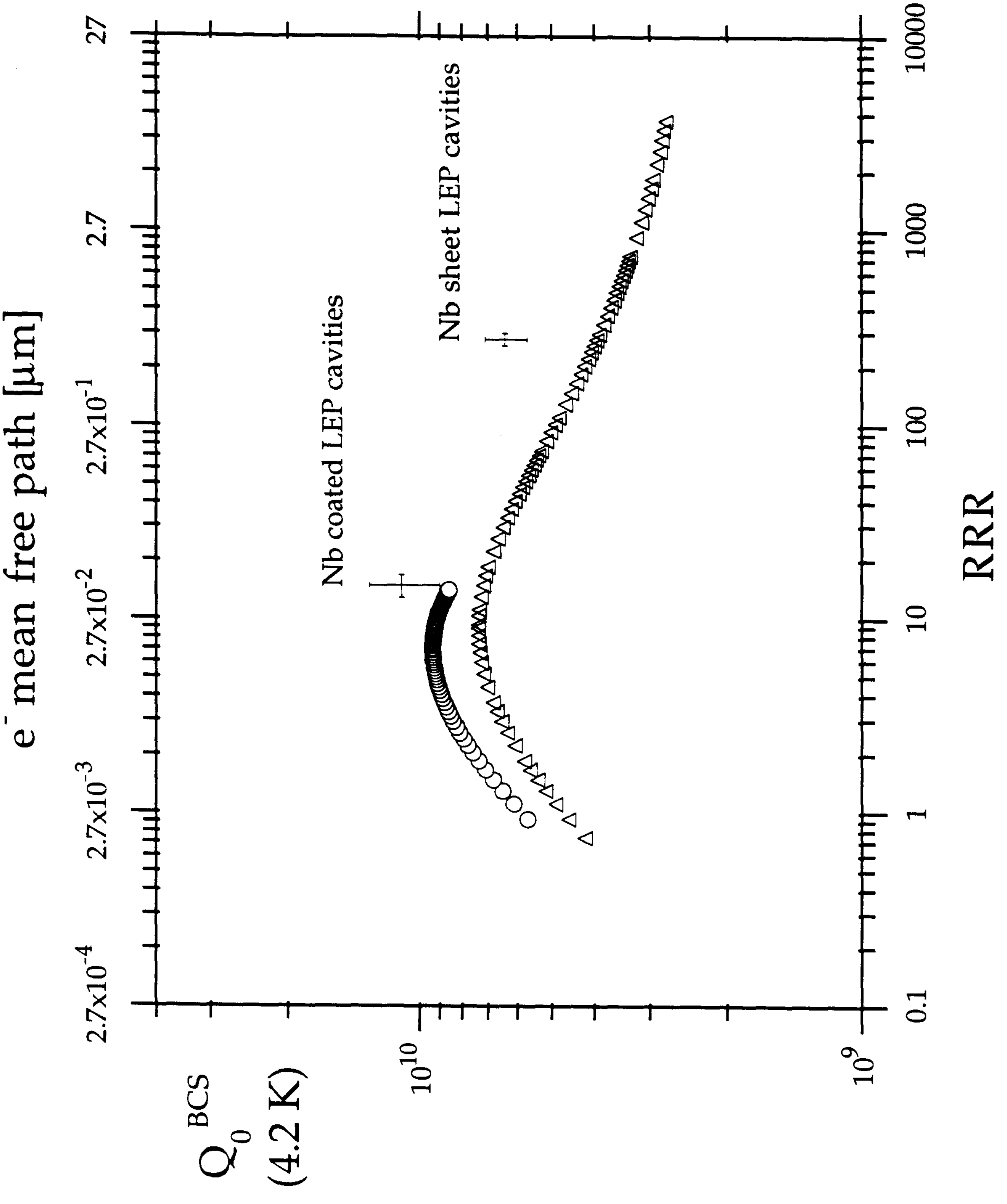
e⁻ mean free path [μm]
2.7x10⁻⁴
2.7x10⁻³
2.7x10⁻²
2.7x10⁻¹
2.7
27
Nb coated LEP cavities
Nb sheet LEP cavities
Q_0^{BCS}
(4.2 K)
10¹⁰
10⁹
0.1
1
10
100
1000
10000
RRR

Fig. 1

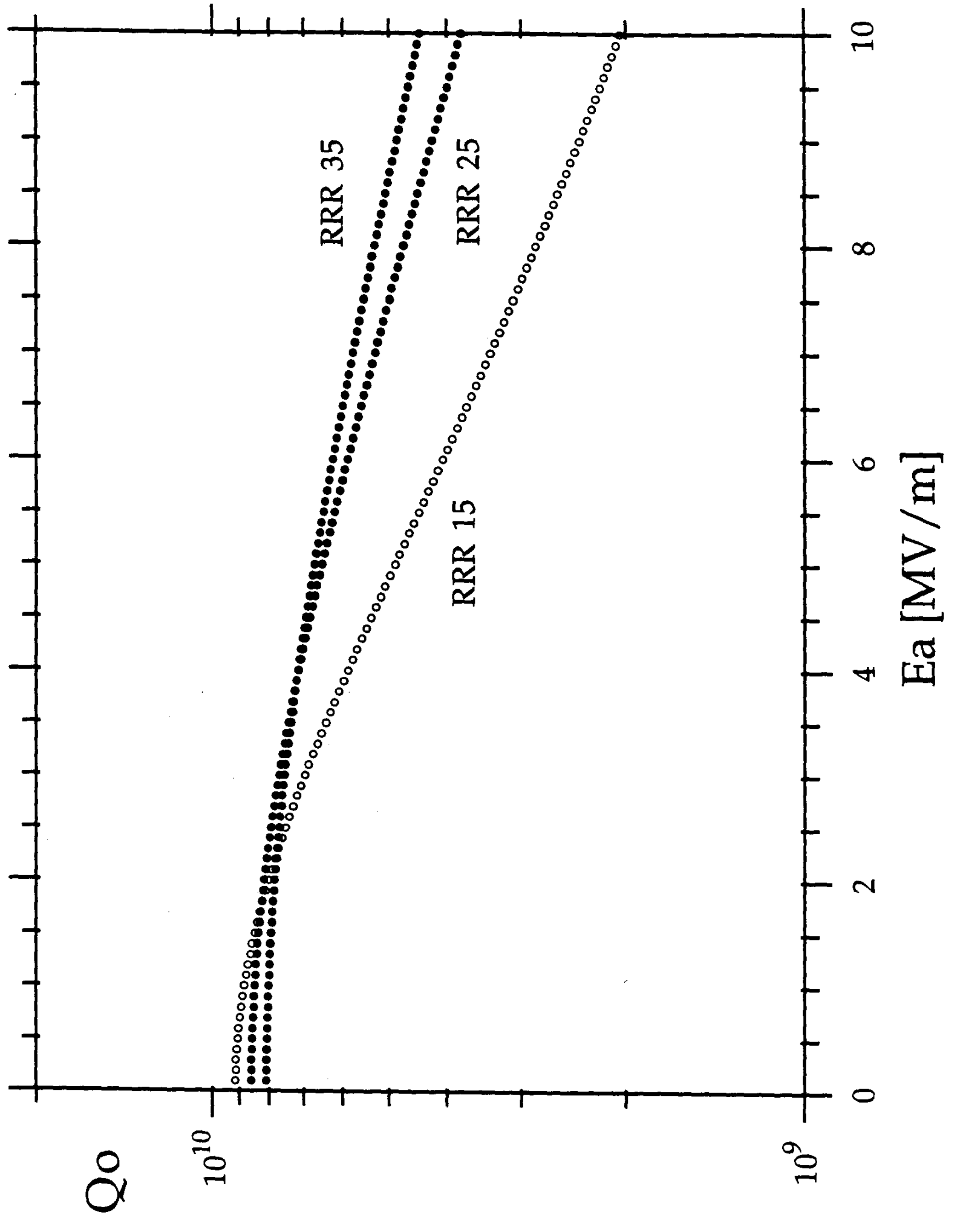

Fig. 2

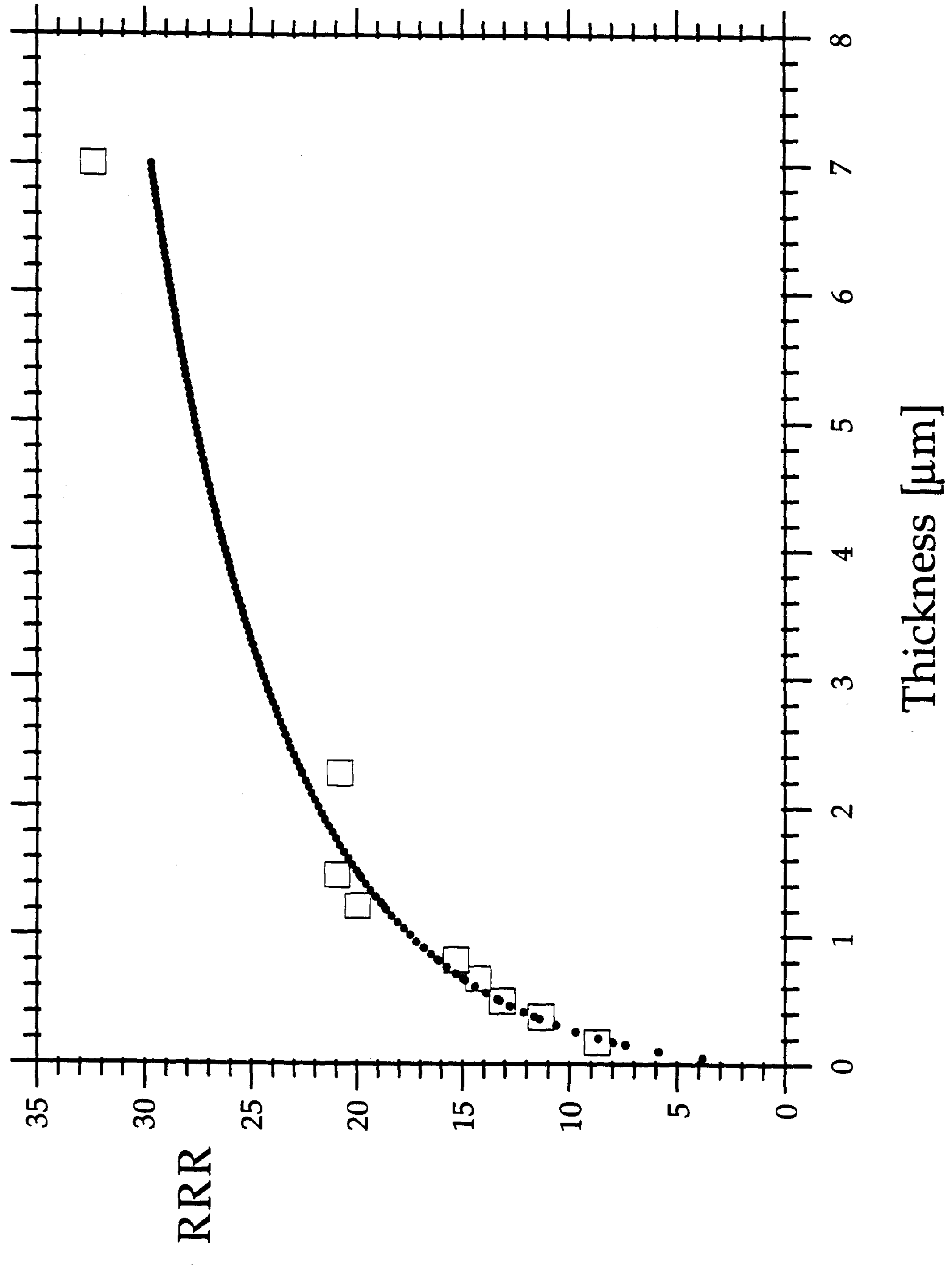

Fig. 3

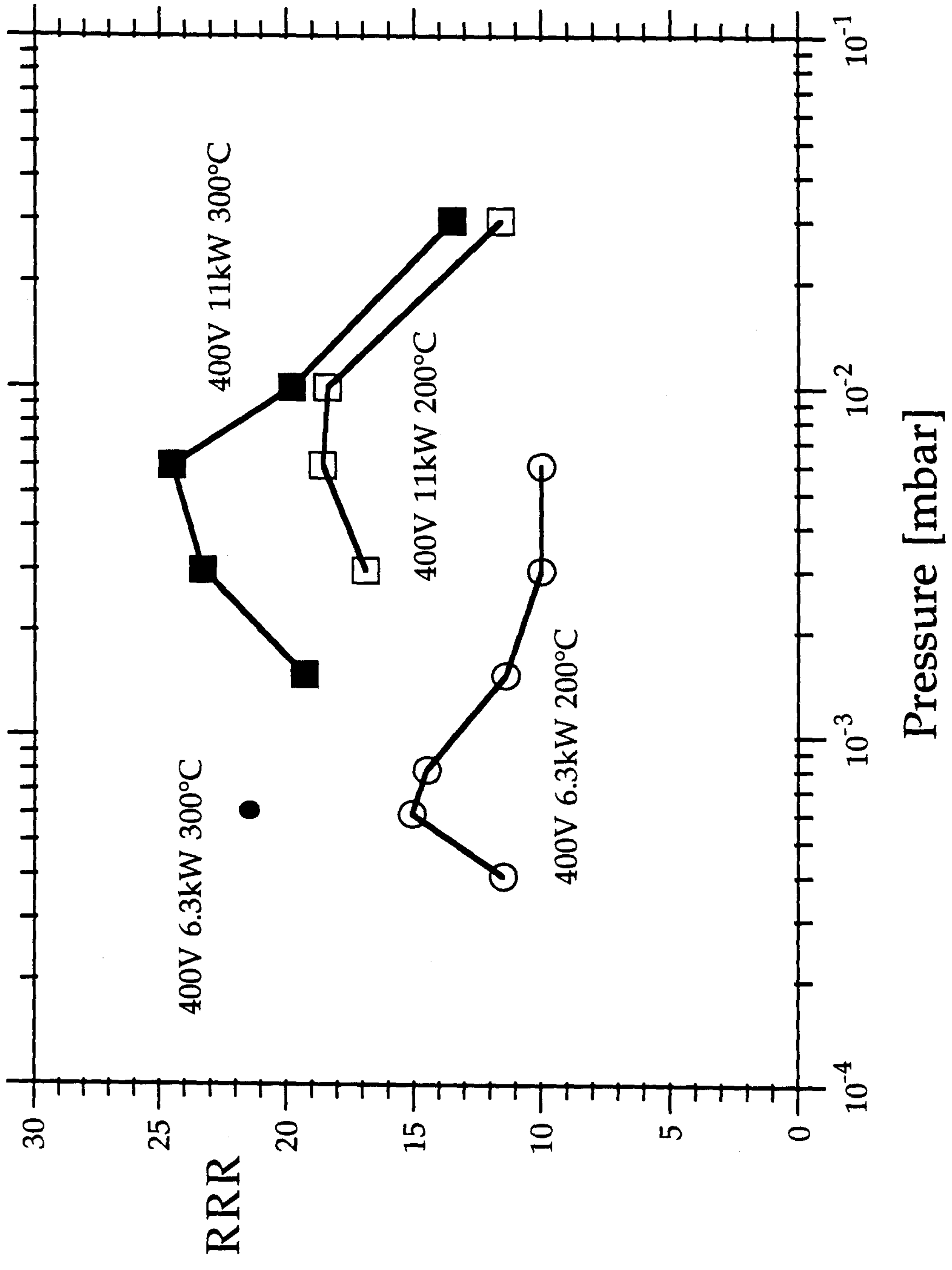
RRR
0
5
10
15
20
25
30
Pressure [mbar]
10⁻⁴
10⁻³
10⁻²
10⁻¹
400V 6.3kW 300°C
400V 11kW 300°C
400V 11kW 200°C
400V 6.3kW 200°C

Fig. 4