CLEANLINESS TECHNIQUES

Detlef Reschke[#], DESY, D-22603 Hamburg, Germany

Abstract

Cleanliness techniques play the key role in the preparation of field emission free, high gradient, low loss superconducting cavities. Contaminations like particles and chemical residues as well as surface irregularities have been identified as major sources of field emission. To avoid these contaminations cleanroom environments and chemical pure, particle filtered media are used.

This paper focuses on the conditions for a clean cavity preparation as well as the discussion of the final processing, cleaning and assembly techniques.

INTRODUCTION

Particles, chemical contaminations like hydrocarbons and surface irregularities have been identified to create field emission. This stresses the importance of the final cleaning and assembly procedures applied to the cavity and its auxiliaries. Moreover particular care has to be taken avoiding any recontamination during the subsequent cavity handling and opration of the accelerator modules. The statement of P. Kneisel and B. Lewis given in the 1995 SRF Workshop [1] still summarizes the situation at its best: "It is generally accepted that the field emission behavior of a niobium cavity reflects the level of cleanliness of the superconducting surfaces subject to the rf-fields."

When applying today's standard preparation procedures, typical field emission loading in well-prepared cavities at 1.3 GHz start at gradients of (20 – 25) MV/m. No systematic degradation between vertical tests and horizontal system performances is found [2] in contrast to older results [3]. Single-cell cavities with their relaxed complexity of necessary components and assembly often achieve gradients far beyond 30 MV/m without field emission [4, 5, 6]. At TTF an electropolished 1.3 GHz nine-cell cavity achieved 35 MV/m without field emission in beam operation [7].

Obviously the cleanliness requirements cannot be confined to the cavity preparation procedures only. The application of well-defined processes during cavity fabrication as well as the appropriate cleaning and handling of all components of the beam vacuum system is necessary.

FIELD EMISSION OF SRF CAVITIES

An overview of srf cavity related field emission effects including dedicated instruments and processing techniques are given in [8,9]. The contributions to the RF Superconductivity Workshops show the historical development starting from 1980 up to now.

[#]detlef.reschke@desy.de

Nature of Field Emitters

This chapter follows closely the respective chapter of ref. [9].

Most field emitters are conducting (metallic) particles of irregular shape with a typical size of $0.5 - 20$ μm (Figure 1). Investigations with dc field emission microscopes and samples in rf fields show that only $5 - 10\%$ of the particles emit. Hydrocarbon contaminations of the surface caused by improper vacuum conditions also result in field emission. Both, dc and rf field emission are well described by the modified Fowler-Nordheim law:

$$I \propto A_{FN} \cdot \frac{(\beta_{FN} E)^2}{\Phi} \cdot \exp\left(-\frac{C\Phi^{3/2}}{\beta_{FN} E}\right) \quad (1)$$

with
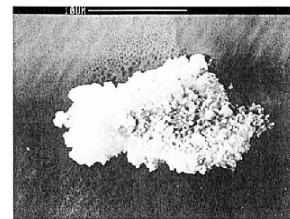|		
	I	emission current
	A_{FN}	Fowler-Nordheim emission area
	β_{FN}	Fowler-Nordheim field enhancement factor
	E	surface electric field
	Φ	work function
	C	constant

Figure 1: Typical emitting particle

The factor β_{FN} gives the local field enhancement at the emitter. Typical observed values vary between 50 and 500. Often emitters with a high β_{FN} can be modified by processing and the β_{FN} decreases resulting in a reduced field emission loading of the cavity. Practically the decrease of the Q-value is shifted to higher fields and the slope is reduced. Usually the emission area A_{FN} is not directly correlated to the physical size of the emitting particle or surface irregularity, which caused controversial discussions about the nature of emission.

With some minor exceptions there is no substantial difference between the dc and rf field emission process.

After long and controversial discussions the present knowledge supports the tip-on-tip model as an appropriate explanation of the experimental observations. One major counter argument against the tip-on-tip model was the field enhancement factor, which does not exceed $10 - 20$

for simple geometric structures. This is in contradiction to the measured values up to a few hundred, both in cavities and on samples. Experiments at Saclay [10,11] resulted in the idea of a nm-scale microtip on top of a μm-sized particle (Figure 2), which explains the observed β_{FN} - values. Within this geometric model adsorbed gases and oxide layers play an important role modifying A_{FN} and β_{FN}. The observed activation of emitters between 200 C and 800 C is explained as a modification of the boundary layer between the substrate (Nb) and the particle together with a modification of the adsorbed gases on the particles. A major role plays the cracking of the isolating Nb_2O_5 - layer and the formation of carbides, sulfides and graphite. Carbon is adsorbed out of the residual gas and sulfur is diffusing out of the Nb. These effects result in an enhancement of β_{FN} by a factor of 2-3 and of A compared to the pure geometrical field enhancement. Firing a Nb surface with emitting particles at > 1200 C renders the surface emission free, which can be argued either by a smoothing of the jagged particle structure or by a strong influence of the interface between niobium and the emitter.

It should be noted that the 120C bake-out [12] is not expected and there is no experimental evidence to effect the field emission behaviour.
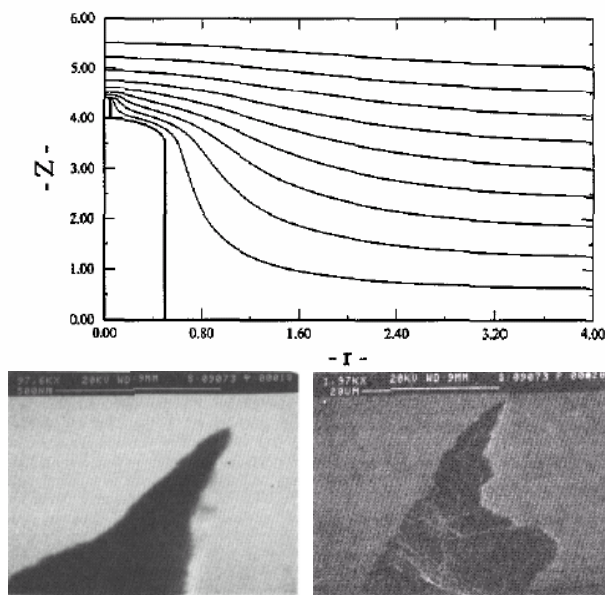

Figure 2: Calculated equipotentials for two superposed hemispherically capped cylindrical projections (top), SEM pictures of sharp emitting structures (bottom) (courtesy of Saclay)

CONTAMINATION AND CLEANING

Each discussion about cleaning techniques first of all requires a detailed specification of the attacked contamination. A simple example may show this. In semiconductor industry one critical contamination are particulates, which shorten the conductor line. In pharmaceutical industry the sterility has highest priority. For cavity applications some critical contaminations are known:

- particles causing field emission
- hydro carbons causing field emission and Q-degradation
- sulphur residues of the electropolishing process may cause field emission

The influence of other contaminations like bacteria in pure water or air/surface molecular contamination (AMC/SMC) on the cavity performance is unknown.

Though a lot of valuable investigations were done in the last years, still the knowledge of the surface conditions of the niobium cavities are poor compared to the standards of semiconductor industry. The complex shape does not allow the application of most of the powerful surface analysis techniques to the inner cavity surface. Investigations of comparably processed samples can compensate this only partially.

A particulate contamination can be chemically dissolved, thermally evaporated or physically removed. The latter is based on overcoming the adhesion forces of the particle at the surface and the subsequent transport out of the cavity. The strength of the adhesion forces depend on a complex mixture of:

- the material, roughness, electrical charge, hardnes of particle and surface
- size and shape of the particle
- temperature and humidity of the environment

Often the van der Waals forces are dominating. As van der Waals forces are weakened in liquids compared to air, wet cleaning processes often show a good effectivity. For hydrophilic materials capillary forces are important. Electric double layer forces play an adhesive role in liquids. The electrical charges form the so-called zeta potential, which depends on the ph-value of the liquid. The zeta-potential is also used for the characterization of particle filters. To make the picture more complicated electrostatic forces and chemical bonds maybe present.

The basics of cleaning technology can be found in dedicated textbooks e.g. [13,14,15]. An excellent discussion of cavity relevant aspects is given in ref. [18].

"STANDARD" CLEANROOM TECHNIQUES

The requirements of design, construction, commissioning and maintenance of high quality cleanrooms and their installations can be found in the industry standards like ASTM, ISO, JIS, VDI, etc.

Proven tools for the quality control are particle counters for air and liquids down to the sub-μm range. Monitoring of the pure water supply systems measuring resistivity, particles, bacteria, TOC, residues and other parameters are standard. Though it is often difficult to correlate the values immediately to the cavity performance, at least the development in time will show upcoming system failures. Recent developments allow the measurement of airborne

particles combined with a spectroscopic analysis and the direct measurement of larger particles on surfaces [16]. Very helpful is the use of a cleanroom compatible airflow visualization. The general flow conditions of the cleanroom, the influence of movement (doors, personal) are visualized as well as special effects at the cavity and its components like open coupler ports (Figure 3) [17].

Figure 3: Flow visualization at a cavity (left) and during movement of personal

CAVITY PREPARATION PROCESSES

The basis for successful cavity preparation has to be provided long before the cleanroom work starts and should be good laboratory practice, but nevertheless is often ignored. The design of all used components must be adapted to cleanroom requirements, i.e. selected materials, design suited for optimal cleaning and handling. A good organisation of the work flow in combination with a well designed cleanroom infrastructure simplifies the activities and avoids unnecessary actions [18]. The processing of each cavity inside and outside of the cleanroom as well as the status of infrastructure has to documented. A complete documentation is essential for cavity data analysis and failure search. The cavity preparation has to be stopped in case of any irregularity, which make a successful cavity test doubtful, and restarted with an adequate cleaning.

The following chapter will sketch the present cleaning and assembly technology. More information can be found in [8,9] and the Workshops on RF Superconductivity.

Ultrasonic cleaning

Ultrasonic cleaning with subsequent DI Water rinse and drying under adapted cleanroom conditions are essential for pre-cleaning and degreasing of cavities and auxiliary components. For cavities it is followed by the final chemical or electrochemical treatment.

Final chemical and electrochemical treatment

Both buffered chemical polishing (BCP) and electropolishing (EP) are no real cleaning techniques, but a removal of a surface layer in µm-range followed by DI-water rinses. The acid mixtures do not remove e.g. organic contaminations like plastics, hydrocarbons, etc., which reveals the importance of an effective pre-cleaning.

The commonly used EP mixture consists of HF and H_2SO_4 in a volume ratio of 1:9. For best removal of hydrogen, produced during the electrochemical reaction, a horizontal set-up is preferred in most labs. The aggressive acid mixture makes high demands on the used materials. Typically fluoric plastics are used for the piping and the electrode is made of Al or Cu. If a copper electrode is used, an additional oxipolishing with HNO_3 and HF is necessary to remove copper traces from the niobium surface [5]. The standard BCP mixture contains HF : HNO_3 : H_3PO_4 in a volume ratio of 1:1:2. Typical for the final treatment is a removal of (10 - 40) µm of the niobium surface. After draining the acid, the cavity is rinsed immediately with water of at least DI-quality. For best removal of acid residues, typically the rinsing is performed in several steps ending with an ultra-pure water rinse ($\rho \geq 18$ MΩcm; particle filtered $\leq 0,2$ µm). Both for BCP and EP closed, PLC controlled systems with integrated rinsing capability for DI or pure water are state-of-the-art (Figure 4). The used acid quality varies, but is often "pro analysi" or better. Additional particle filtration is often integrated in the chemical system.

Open questions concern the level of acid quality and particle filtration. Alternative mixtures have been investigated in the past, especially BCP with a volume ratio of 1:1:1 showed excellent results. The EP mixture shows dramatic HF degassing, which requires adequate precautions and handling of the process. This effect needs further investigation. Hot water rinsing after the acid draining should result in a better solubility of residues and better drying. On the other hand undesirable reactions may start at the surface. The required cleanliness of the preparation environment ("good" lab standard vs. cleanroom cl. 10.000 or better?), especially of large scale production, is not finally settled. The implementation of complex chemical and mechanical process equipment, especially for EP, in a cleanroom requires advanced technical solutions.

Figure 4: Closed BCP facility at JLab (top left, courtesy of Jefferson Lab), EP facility at Nomura Plating (top right, courtesy of Nomura plating), EP facility at DESY (bottom left), BCP facility at DESY (bottom right)

High Pressure Rinsing

At present repeated rinsing with high-pressure ultra-pure water (HPR) is the most effective tool to avoid field emission loading. Typically, HPR systems (Figure 5) work with a water flow between 5 l/min and 20 l/min and a pressure between (80 - 150 bar), which allows removal of particles larger than a few micrometer [1]. To avoid any recontamination, the cavity is rinsed in a cleanroom environment of cl. 100 or better, in a glove box or is closed with protection flanges. Repeated rinses are advantageous in order to rinse out particles, which have been loosened off the cavity surface, but depending on the water flow conditions have been transported and re-deposited inside the cavity. Experience at DESY showed that it is important to avoid drying before starting the first rinse. A possible explanation is that after drying particles stick stronger to the surface and removal becomes more difficult.

The technical installations like pump, piping, turntable and nozzle system differ widely and thus are not described. It only should be stressed, that the final particle filter (pore size $\leq 0,2$ µm) has to be placed as closely to the nozzle as possible with no moving parts (i.e. valves) or dead ends between filter and nozzle. Furthermore no parts, which come in contact with bearings etc, should be moved inside the cavity. A design with fixed cane, enclosed bearings and all moving parts far off the open cavity is preferable.

Quality control aspects of HPR systems are twofold. Provided that the water system produces the desired pure water quality, the high pressure pump, valves and filter units can act as sources of contaminations (particles, hydrocarbons) of the high-pressure water in case of a component failure. An on-line measurement with respect to the above mentioned contaminations is highly desirable and first realized at JLab [19]. Secondly, it was tried to monitor the cleaning effect by measuring the particles rinsed out of the cavity using a particle counter [20] or a filter [21,22]. During the first HPR after BCP or EP a large amount of particles is rinsed out of the cavity. All materials used in the cavity preparation like rubber, copper, steel and even large particle up to >100µm are found. In subsequent rinses the particle number decreases drastically. Though these measurements give valuable information about the rinsing and cleaning effect, up to now no conclusive correlations to the cavity performance are found. New clever and applicable ideas (e.g. particle concentration on a Nb sample) are needed.

The option of additional outside rinsing maybe helpful to avoid contamination transport from the chemistry area to the cl.10/100 assembly area. This holds especially for multi-cell cavities with their complex shape. Theoretically a higher pressure than (100 – 150) bar results in a reduced size of removable particles, which decreases inversely to the square root of the pressure. Reported investigations on the damage of the niobium surface are rare [20] and indicate, that a gain of 30 – 40 % reduction in particle size

can be achieved theoretically. First investigations of the influence of the jet parameters on the cleaning force have been started recently [23].
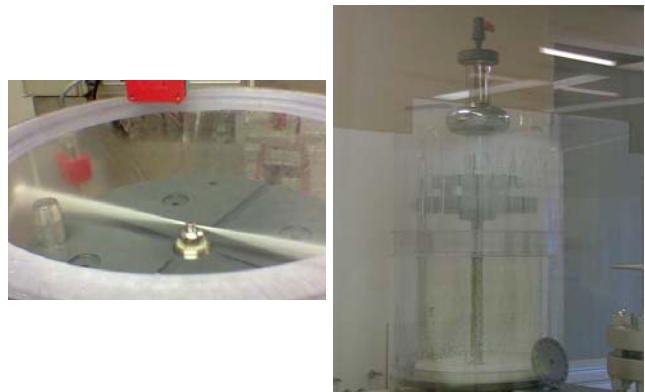

Figure 5: HPR stand at CEA Saclay (right, courtesy of CEA Saclay), HPR jets at 150 bar (Left)

Pumping and Venting

Oil-free pump stations equipped with helium leak detector and residual gas analyzer are state-of the art to avoid any risk of a hydrocarbon contamination. More details are described in [9,24]. Venting is done using pure, dry and particle filtered nitrogen or argon gas. Laminar venting prevents particle transport due to turbulences in the pump line and cavity (Figure 6) [25].
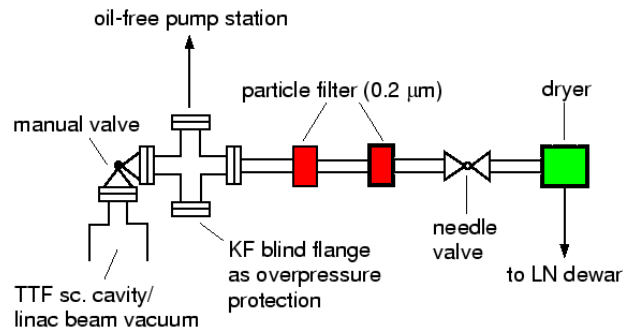

Figure 6: Schematic layout of the set-up used for venting of cleaned vacuum systems at TTF

The influence of storing a high performance cavity using typical gases like argon, nitrogen or cleanroom air, is not fully explored yet and different investigations came to contradicting results [7,26].

Assembly and Drying

As mentioned above, the essential conditions for a contamination-free assembly are given by the design of all involved components long before the cleanroom actions start. Especially the flange connections and the gaskets attached to the cavity, which necessitate an easy handling as well as a reliable leak tightness, are of outstanding importance (Figure 7) [24].
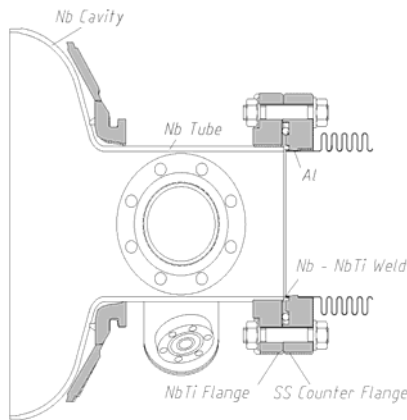
Figure 7: Flange design of the TTF cavities using NbTi-flanges and massive aluminum gaskets

After cleaning and drying, the cavity and its components are assembled in a cleanroom environment better than class 100. Blowing off both, the components and tools, with pure ionized gas immediately before the assembly in front of a particle counter can be used as a good check for the particle contamination as well as a final removal of remaining particles. This is of particular importance during the final assembly of a cavity or the connection of cavities before beam operation, where no cleaning can be applied afterwards. It is evident, that the handling and assembly time at an open cavity should be as short as possible. Finally, best design and cleaning will not help, if the cleanroom staff is not well trained and highly motivated.

Alternative measurement techniques of surface contamination may become helpful tools to improve the assembly reliability.

Finally the general avoidance of bolted flanges with their high risk of particle contamination should be mentioned. Though there are many unsolved questions, like for example how to realize an ultrapure welding procedure under cleanroom conditions, this option is it worth to be investigated further, especially for large scale applications. First tests with two 1.3 GHz seven-cell cavities connected by electron beam welding to a "superstructure" were successful [27].

Various applied drying procedures are discussed in [9]. Due to missing systematic investigations, no assessment of drying procedures can be given.

Furnaces

Today's cavity preparation includes at least an 800°C firing under vacuum conditions. Typically the furnace is not integrated in the cleanroom infrastructure, which at least results in a particle contamination of the surface and the necessity of an additional cleaning, e.g. ultrasonic cleaning + pure water rinse. For an optimized workflow the integration in the cleanroom environment e.g. loading of the furnace close to the chemistry area (cl. 1000 – cl.10000), seems to be reasonable.

A furnace for the high temperature (1200 – 1400)°C postpurification process is part of the cl.100 area of the proven TTF preparation infrastructure.

Tuning and Inspection

Mechanical and optical inspections as well as tuning of the cavities are important steps of the preparation process. With the increasing requirements of cleanliness and for an optimized workflow, again the integration of these procedures in the cleanroom infrastructure avoids needless contamination and cleaning. Obviously tuning and inspection requires complex mechanical and optical equipment, which is not suitable for cl.100 or better. On the other hand the need of new well-designed equipment usable under cleanroom conditions of e.g. cl.1000 should not prevent a substantial improvement of the overall preparation procedure.

Alternative CleaningTechniques

Following the requirements of semi-conductor industry a number of advanced cleaning techniques have been developed for smooth wafers [1,13,14,15]. Due to the complex shape of the inner surface most of them are not applicable to cavities. After first considerations and pilot tests only megasonic and dry-ice cleaning seem to have potential for cavity cleaning.

The principle of megasonic cleaning is similar to ultrasonic, but with frequencies around 1 MHz. The cleaning effect is based on high power pressure waves inside the cleaning solution less than on cavitation. Particles down to 0,1 μm can be removed from wafer surfaces. First cavity results showed promising results [28], but also the need to develop an oscillator applicable inside the cavity to realise a high transmission of megasonic power. The transportation of particles out of the cavity requires a high flowrate, which is no problem for an open cavity, but might need some technical effort for cavities with assembled flanges.

Dry-ice cleaning with CO_2-snow allows effective cleaning of sub-micron particles and film contamination by a combination of mechanical, thermal and chemical effects (Figure 8). The cleaning process acts local, mild, dry, without residues and requires no additional cleaning agent. Cleaning of niobium samples and first cavity tests show promising results [29]. As the particle transport is based on a gas flow out of the cavity, horizontal cleaning of cavities seems to be possible in contrast to HPR. Furthermore, the dry cleaning would preserve the effect of preconditioning of a rf power coupler attached to a cavity.

Figure 8: Test of the dry-ice nozzle system in a cut NbCu-cavity

SUMMARY AND OUTLOOK

Today's standard cleaning, handling and assembly procedures often allow an excellent cavity performance meeting the requirements of the next accelerator projects. Nevertheless field emission, resulting in undesirable dark currents, is still the main limitation, if usable gradients above 20 MV/m are required. Therefore further improvements of the standard preparation procedures as well as the development of alternative approaches are necessary. A decisive role plays the further development of efficient quality control procedures.

Of highest importance for a reliable preparation is an effective quality control and assurance of the HPR system. This contains the measurement of the critical parameters of both the low pressure ultrapure water system and, more difficult, the high pressure line close to the nozzle system. A practical approach to judge about the cleaning efficiency is needed. Checking the particles of the drained water coming out of the cavity gives valuable information, but additional new clever ideas are necessary.

For upcoming large scale applications with high gradient requirements it is of outstanding importance to simplify components and procedures with respect to an optimized work flow under cleanroom conditions.

Today' knowledge supports electropolishing combined with a thorough HPR treatment (see above) as final surface preparation in order to achieve high gradients. Nevertheless the EP process is not sufficiently understood. "Q-disease" caused by hydrogen pollution cannot be avoided reliably. The HF degassing and the stability of the acid mixtures requires more investigation. The design of an EP system capable for reliable large-scale operation needs refinement.

A cleaning option of the horizontal, fully equipped cavity before the final module assembly seems to be helpful to preserve high gradients up to the accelerator operation. Dry-ice cleaning may have substantial benefit and shows promising first results.

REFERENCES

1 P. Kneisel, B.Lewis, Proc. of 7th Workshop on RF Superconductivity, Gif sur Yvette, France (1995), p.311

2 Editors: R. Brinkmann, K. Flöttmann, J. Rossbach, P. Schmüser, N. Walker, H. Weise; TESLA Technical Design Report, DESY 2001 - 011, ECFA 2001 - 209, TESLA Report 2001 - 23, TESLA - FEL 2001 - 05 (2001)

3 T. Furuya, "Preparation and Handling of Superconducting RF Cavities", Proc. 4th Workshop on RF Superconductivity, Tsukuba, Japan, p. 305 (1989)

4 M. Ono et al., "Achievement of 40 MV/m Accelerating Field in L-Band SCC at KEK" and K.Saito et al., "Superiority of Electropolishing over Chemical Polishing on High Gradients", 8th Workshop on RF Superconductivity, Abano Terme, Italy, p.472, 1997

5 L.Lilje et al., "Improved surface treatment of the superconducting TESLA cavities", Nucl. Instr. and Meth. 516 (2-3), (2004), 213

6 R. Geng et al., „High Gradient Activities at Cornell: Reentrant cavities", this conference

7 L.Lilje et al. "Achievement of 35 MV/m in the superconducting nine-cell cavities for TESLA ", Nucl. Instr. and Meth. 524 (1-3), (2004), 1

8 H. Padamsee et al.; "RF Superconductivity for Accelerators"; John Wiley & Sons Inc., ISBN 0-471-15432-6, 1998

9 D.Reschke, "Field Emission Overview: Cleanliness and Processing", Proc. of the Workshop on Pushing the Limits of RF Superconductivity, Argonne, ANL-05-10 (2005)

10 M. Jimenez et al., J. Phys. D, Appl. Phys., 27: 1038 (1994)

11 R. Noer, Proc. 6th Workshop on RF Super-conductivity, CEBAF, Newport News, USA, p.236, (1994)

12 B. Visentin, "Q-Slope at High Gradients: Review of Experiments and Theories", Proc. of 11th Workshop on RF Superconductivity, DESY, Luebeck, Germany

13 D. L. Tolliver ed., "Handbook of Contamination Control in Microelectronics", Noyes Publications, ISBN 0-8155-1151-5, 1988

14 W. Kern ed., "Handbook of Semiconductor Cleaning Technology," Noyes Publications, ISBN 0-8155-1331-3, 1993

15 L.Gail, H.P.Hortig, Reinraumtechnik (in german), ISBN 3-540-66885-3, 2002

16 Th. von Kahlden, private communication, www.cci-vk.de

17 N. Krupka," Quality Control Update of the Cleanroom for Superconducting Multicell Cavities at DESY", this conference

18 K. Escherich et al., "Clean Room Facilities for High Gradient Resonator Preparation", this conference

19 J. Mammosser, "Update on Cleanroom, Clean Water, HPR and Field Emission", SMTF Collaboration Meeting, Oct 5-8th, FNAL, USA, http://ilc-dms.fnal.gov/Workgroups/SMTF/

20 D. Bloess, CERN, private communication

21 J. Delayen et al., Proc of the LINAC 2004, Luebeck, Germany, p.226 (2004)

22 D. Reschke, "New Aspects of Quality Control during Preparation of TTF 1.3GHz Cavities", Proc. of the 9th Workshop on RF Superconductivity, Santa Fe, USA, p.159, 1999

23 P. Michelato, "High Pressure Rinsing Parameters Measurement and Process Optimization", this conference

24 K. Zapfe-Düren et al., "A New Flange Design for the Superconducting Cavities for TESLA", 8th Workshop on RF Superconductivity, Abano Terme, Italy, p.457, 1997

25 N. Krupka, "Quality Control at the TTF Cleanroom Infrastructure for Cavity Procesing", Proc. of the 10th Workshop on RF Superconductivity, KEK Proc. 2003-2, Tsukuba, Ibaraki, Japan (2003)

26 K. Saito, Proc. of the 10th Workshop on RF Superconductivity, KEK Proc. 2003-2, Tsukuba, Ibaraki, Japan (2003) p.423

27 J. Sekutowicz et al., "Status of the Superstructure", Proc. of the 10th Workshop on RF Superconductivity, KEK Proc. 2003-2, Tsukuba, Ibaraki, Japan (2003) p.177

28 K. Saito et al., "Water Rinsing of the Contaminated Superconducting RF Cavities", Proc. 7th Workshop on RF Superconductivity, Gif sur Yvette, France, 1995, p. 379

29 A. Brinkmann et al., "Further Improvements with Dry Ice Cleaning on SRF Cavities", this conference