Study of Ultra-clean Surface for Niobium SC Cavities

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Abstract

For a TeV energy particle physics, R&D of electron/positron linear colliders has been conducted very hard at many laboratories. Development of high field gradient cavities is one of key issues to realize such a machine. Field emission loading is a dominant phenomenon limiting high field. Particulate contamination on the inner surface is a main source of field emission. In this paper, we report about particulate contamination in wet surface treatments. We discuss particles in chemicals and contamination from environments. Studies with high pressure ultrapure water rinsing and megasonic rinsing are also described.

I. INTRODUCTION

Niobium material with high thermal conductivity suppressed the thermal instability and improved the field gradient over 10 MV/m in superconducting cavities. Today field emission loading is a main obstacle which limits the maximum field in superconducting niobium cavities. Particulate contamination is a main source of the field emission. Therefore, many efforts have been performed to eliminate these seeds with surface treatments. Especially, in the last SRF workshop at DESY, CERN people reported a promising result on high pressure ultrapure water rinsing against field emission [1]. Since the last workshop, such a cleaning technique is applied extensively in many laboratories and an electric surface peak field (Ep) over 50 MV/m is achieved reproducibly without field emission loading [2]. On the other hand, Cornell people have developed high peak power processing as an in-situ cleaning method. They have obtained the accelerating field gradient (Eacc) of 25 MV/m in a 1.3 GHz five-cell cavity [3]. These complementary approaches are getting a final goal of TESLA-500; Eacc=25 MV/m without field emission loading.

KEK also is progressing in high field [4]. We have applied the TRISTAN's traditional careful rinsing [5] to L-band cavities. Field emission loading is not a major problem as to our L-band single-cell cavities. We have achieved the maximum field gradient of 30 MV/m without field emission loading. Our current field limitation is a fast breakdown and the Japanese Q-disease [6]. We have two reasons to continue clean work; one is to improve reliability in our surface preparation, and the second is a cost down of the surface treatment. KEK's rinsing method takes a long time about 3-5 hours. We want to establish an effective and speedy rinsing method. If possible, we wish to cure the present our field limitation by this study. As the first investigation, we estimated the particle contamination from environment, and evaluated the dust particles in chemicals which we use usually in cavity preparation. Then we examined particles on the surface to know about our present rinsing state. We tried high pressure ultrapure water rinsing to our cavities and investigated its effect on the cavity performance. In addition, we tested megasonic rinsing to samples, and made sure its large ability against particulate contamination. In this paper we will describe results of these investigations.

II. EXPERIMENTS

First of all, we counted the particle number in chemicals with a particle counter for liquids; Particle counter KL-22 from the Rion Co. Ltd.. The investigated chemicals are sulfuric acid (H_2SO_4), hydrofluoric acid (HF), phosphoric acid (H_3PO_4), nitric acid (HNO_3), ethanol, methanol, isopropyl alcohol, ultrapure water and pure water. These are often used in the preparation of superconducting niobium cavities. We always sampled every chemical from a new bottle. The ultrapure water and pure water were sampled from our surface treatment system. We measured two kinds of chemical grades; reagent grade (R-G) and electric grade (EL-G).

Next, we measured the number of particles on surfaces of samples that were treated with various rinsing methods; TRISTAN rinsing method [5], high pressure ultrapure water rinsing (HPR, 85 kg/cm^2), ultrasound agitation (28, 100KHz), and megasonic rinsing (950 KHz). We adopted a same equipment as Scalay [7] to count particles. Namely we used a scanning laser particle counter. Semiconductor people use this equipment to evaluate particles on silicon wafers. The principle of the measurement is; a laser beam scans on a silicon wafer, and a scattered light signal from an eventual particle on the surface is amplified with a photo multiplier. This equipment provides us information about the number of particles and its distribution on the surface (see Fig.6,7).

We employed silicon wafers (100$^{\Phi}$, 0.5^{t}, N-type) with a smooth and clean surface from the Shinetsu Kagaku Company. Wafers were treated at the Nomura Company and sent to the SONY Technology Center for analysis. We used a vacuum pincette to handle wafers. Fig.1 shows the setup for the rinsing experiment. As we used the clean room (CR) as a stockroom for surface treatment tools, the cleanness would be worse than class 100. We took a small class 100 clean booth (CB) into the clean room. Works such as putting in and out of wafers from containers, inspection by eye, drying and so on were performed in this clean booth. Fig.2 shows an

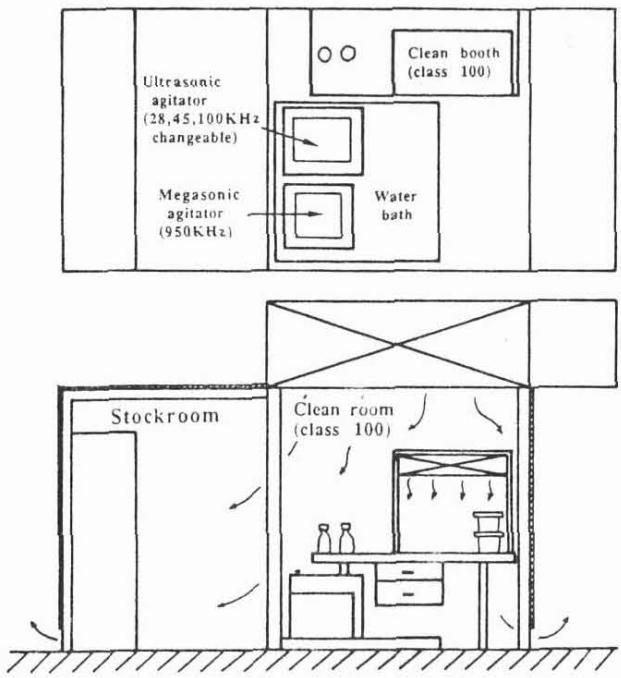


Fig. 1 A setup for the rinsing experiment.

Fig. 2 Surface inspection in the clean booth.

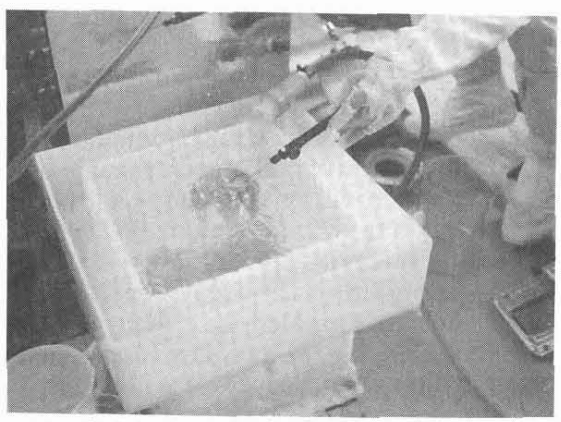


Fig. 3 Megasonic rinsing in the clean room.

inspection scene of silicon wafers in the CB. After inspecting wafers in the CB, they were treated in the CR according to procedures summarized in Table 1, where "Rinse" means that ultrapure water is simply poured on wafers. Fig.3 exhibits the megasonic rinsing in the CR. Only in the case of HPR, we used a divisible cavity (Fig.4). This cavity can contain four wafers, which are pushed on holders by atmospheric pressure. Fig.5 shows our HPR system for L-band single-cell cavities. Ultrapure water from our ultrapure water system is pressurized up to 85 kg/cm^2 by a diaphragm pump. We used electropolished clean stainless steel pipes for the tubing in this system. Ultrapure water jets on the cavity inner surface through the final filter (0.2 μm) at the inlet of the cavity.

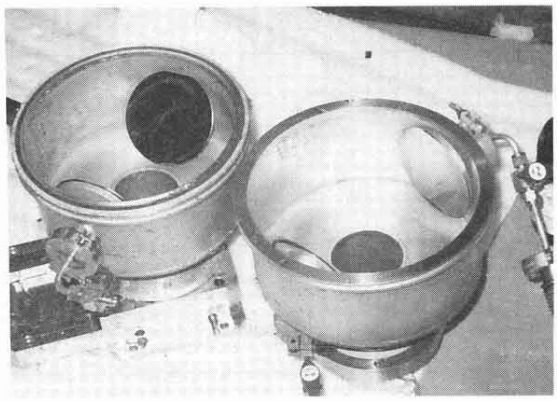


Fig. 4 A half divisible cavity to set silicon wafers inside.

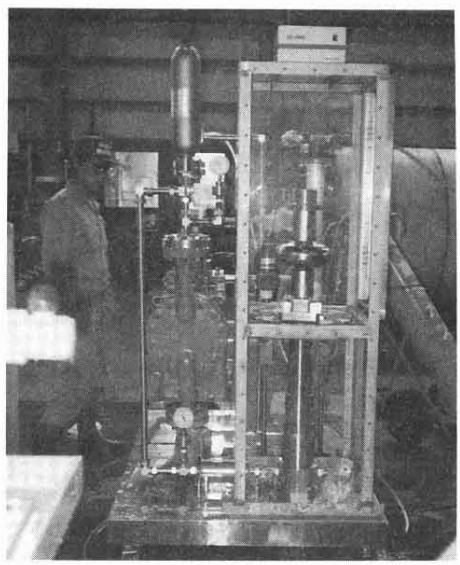


Fig. 5 High pressure ultrapure water rinsing system for L-band single-cell cavities.

III. RESULTS AND DISCUSSION

A. Dust particle from working environment or chemicals

Although the numbers of particles are scattered in some cases (see Table 1), we can roughly summarize as follows.

1) Usual environment is dangerous source of particles, it brings few tens of thousands in 10 min..
2) Contamination in a nominal class 100 CR looks little enough if exposure is short, and it can be reduced to much less than half if CR is used carefully.
3) Wet surface is liable to trap particles, so wet treatment should be done in a clean environment.
4) HPR is better than ultrasound agitation but the best result is given with megasonic rinsing.
5) Contamination from chemicals is little enough, even if R-G sulfuric acid is used.

B. Particles in chemicals

We report the results on particle measurement in chemicals and water in Table 2. The numeral in the table is the number of particles in 10 ml, averaged value in ten measurements. Its standard deviation is also noted by ±. Usually EL-G guarantees that particles larger than 0.5 μm are eliminated less than 1000 pieces in 10 ml for acids, and 500 pieces for solvents. Volume or weight in Table 2 means a difference in bottle size. The remarkable results are ;

1) Hydrofluoric acid, Ethanol and Isopropyl alcohol, which are very important chemicals in semiconductor technology, are well controlled in EL-G on the dust particles larger than 0.3 μm. Especially for ethanol and hydrofluoric acid, even in the reagent grade particles are comparably less to the electric grade.
2) For nitric acid and phosphoric acid which we are using now, particle control is loose comparing to the chemicals mentioned above.
3) Our ultrapure water is well controlled against particles larger than 0.3 μm.

Table 1 Rinsing conditions and the residual particle number on wafers.

Treatment	No.	Condition	Integrated particle number (μm)
Background	1	in CB for 12 hr.	238, 892 (≥0.27) 166, 521 (≥0.30)
	2	in CR for 12 hr.	3265, 4010 (≥0.27) 940, 991 (≥0.38)
	3	2 → Outside CR for 10 min.	18819 (≥0.27)
Water rinsing	4	Rinsed for 1 min. in CR	645, 1864 (≥0.27)
	5	Outside CR for 10 min. → Rinsed for 10 min. in CR.	10254 (≥0.27) 7929 (≥0.30)
Immersed in H_2SO_4	6	Immersed to EL-G H_2SO_4 for 5 min. in CB. → Rinsed in CR for 5 min.	67, 6967 (≥0.30)
	7	Immersed to R-G H2SO4 for 5 min. in CB. → Rinsed in CR for 5 min.	227, 575 (≥0.30)
	8	Immersed outside CR to R-G H2SO4 for 5 min. → Rinsed outside CR for 5 min. Rinsed for 1 min. in CR.	407, 2177 (≥0.30)
Ultrasound agitation 28,100kHz	9	6 → 28kHz for 10 min. in CR. → Rinsed for 1 min. in CR.	2050, 5125 (≥0.30)
	10	7 → 28kHz for 10 min. in CR. → Rinsed for 1 min. in CR.	1516, 1808 (≥0.30)
	11	8 → 28kHz for 10 min. in CR. → Rinsed for 1 min in CR.	2573, 2670 (≥0.30)
	12	8 →100kHz for 10 min. in CR. → Rinsed for 1 min. in CR.	1498, 2342 (≥0.30)
Megasonic rinsing 950kHz	13	6 →950kHz for 10 min. in CR. → Rinsed for 1 min. in CR.	712, 747 (≥0.30)
	14	7 →950kHz for 10 min. in CR. → Rinsed for 1 min. in CR.	110, 128 (≥0.30)
	15	8 →950kHz for 10 min. in CR. → Rinsed for 1 min. in CR.	1179 (≥0.30)
	16	Outside CR for 2 hr. → 950kHz for 10 min. in CR. → Rinsed for 1 min. in CR.	224 (≥0.30)
HPR 85kg/cm^2	17	6 → HPR for 10 min. → Rinsed for 1 min. in CR.	2709 (≥0.30)
	18	7 → HPR for 10 min. → Rinsed for 1 min. in CR.	1017 (≥0.30)
	19	Outside CR for 2 hr. → HPR for 10 min. → Rinsed for 1 min. in CR.	1118 (≥0.30)

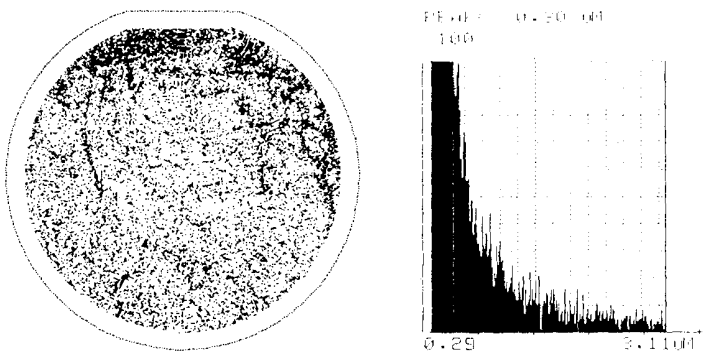
Rinsed; Ultrapure water is simply poured on the wafer.

Table 2. The number of particles in chemicals of 10 ml.

Particle size / Chemicals	0.3 - 0.5 μm	0.5 - 1.0 μm	1.0 - 2.0 μm	Total
H_2SO_4, R-G 95 %, 500 ml	902 ± 24	98 ± 9	17 ± 5	1019 ± 19
H_2SO_4, R-G 95 %, 15 kg	13285 ±329	1006 ± 17	86 ± 7	14388 ± 322
H_2SO_4, EL-G 96 %, 1 kg	2952 ± 85	240 ± 14	40 ± 3	3252 ± 88
HF, R-G 46 %, 500 g	617 ± 15	230 ± 19	41 ± 6	898 ± 22
HF, R-G 46 %, 25 kg	8 ± 4	0	0	8 ± 4
HF, EL-G 50 %, 1 kg	8 ± 2	0	0	8 ± 2
H_3PO_4, R-G 85 %, 25 kg	11333 ± 398	864 ± 76	117 ± 16	12335 ± 402
HNO_3, R-G 61 %, 500 ml	8061 ± 40	578 ± 9	40 ± 3	8683 ± 50
HNO_3, R-G 60 %, 25 kg	42158 ± 155	7682 ± 74	513 ± 33	50374 ± 47
Ethanol, R-G 500 ml	180 ± 3	8 ± 2	1 ± 1	190 ± 4
Methanol, EL-G 1000 ml	291 ± 6	14 ± 3	1 ± 1	307 ± 8
Isoplopil Alchole R-G	5642 ± 62	724 ± 50	26 ± 6	6394 ± 47
Isoplopil Alchole EL-G	58 ± 14	9 ± 2	1 ± 1	68 ± 13
Ultrapure water KEK inline	11 ± 5	1 ± 1	0	12 ± 5
Purewater KEK (10 MΩ cm)	39178 ± 2731	1782 ± 230	131 ± 23	41099 ±2974

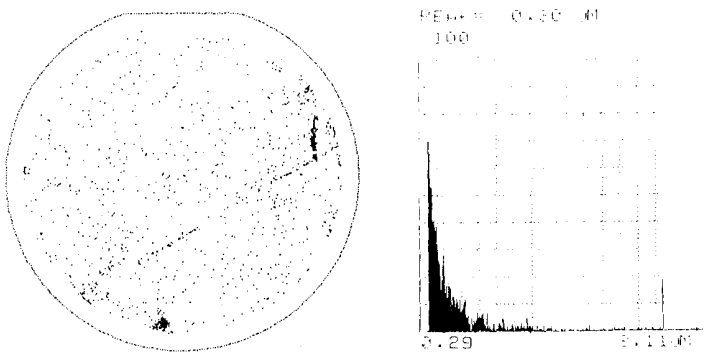
C. Residual dust particles in the TRISTAN rinsing method

Currently a field emission loading is observed in a 508 MHz single cell cavity treated with the TRISTAN rinsing procedure[8]. We simulated the rinsing method with silicon wafers for its cure and evaluated the residual particle on the surface. We exposed wafers to our ordinary working environment for 10 minutes; outside the CR, then immersed them in the electropolishing acid (H_2SO_4[97%]:HF[46%] = 10:1 in volume, R-G) for 5 minutes outside the CR. Wafers are slightly etched by the acid. We could not observe its influence in brightness measurement but detected the weigh reduction by 0.15 %. We rinsed wafers for 5 minutes in the CR with ultrapure water (first rinsing), then attached them in the divisible cavity. Successively, we took a "shower rinsing", hydrogen peroxide (H_2O_2) rinsing, and final overflow rinsing in a 50 °C hot bath. We took samples in each process and measured particle number with the scanning laser particle counter. At the initial stage, the particles (> 0.3μm) of more than 14000 pieces cling on the surface; the counter overflowed due to too much contamination. After the shower rinsing, the number reduced to 13000, and was almost same, 13250, after H_2O_2 rinsing. Fig. 6 is a result after the final rinsing. Fig.7 is a result for a wafer rinsed with HPR after the "shower rinsing". In these figures, the number of 1.2-2.01 μm particle is unnaturally smaller than that expected from the other numbers. This is due to the less sensitivity of our particle counter. Anyway still 10000 pieces left on the surface after the final rinsing. If we use HPR, particles reduce to 1000. Once the surface gets dry, it is difficult to remove particles. For example, when we rinsed again with HPR the same wafer which was kept clean after the measurement of Fig.6, the particles decreased only from 10000 to 8500, and to 2650 with the megasonic rinsing. In this experiment, the etched effect of wafers brings an ambiguity on the absolute number of particle but we believe that the qualitative tendency dose not change.



Particle size	Count
0.30-1.20 μm	5825
1.20-2.01 μm	405
2.01-3.00 μm	2720
> 3.00 μm	1069
Total	10019

Fig. 6 Residual particles on a wafer surface after the TRISTAN final rinsing.



Particle size	Count
0.30-1.20 μm	646
1.20-2.01 μm	52
2.01-3.00 μm	282
> 3.00 μm	37
Total	1017

Fig. 7 Residual particle on a wafer surface after HPR.

D. Effect of HPR

In the last SRF workshop at DESY, a very impressive result on HPR was reported from CERN [1]. This result is about niobium coated copper cavities but that is expected to be valid to niobium bulk cavities. Therefore we developed a HPR system shown in Fig.5. At first, we investigated HPR effect on residual particle on the surface with this system. We applied HPR to silicon wafers with a solid nozzle (see Fig.8) for 10 minutes at the pressure of 85 kg/cm^2. Comparing Fig.7 with Fig.6, one can see a clear effect in HPR. HPR enables to eliminate up to one-tenth of the TRISTAN's.

Next we applied it to L-band single-cell cavities. In this experiment we used two kinds of nozzle; a flat nozzle of which chip is made of tungsten carbide, and a solid nozzle made of stainless steel (see Fig.8). Fig. 9 compares results of cavity performance with HPR (85 kg/cm^2, 40 min.) and our standard rinsing. Here, C-2 cavity (mark; ●) was treated with the flat nozzle in HPR, and C-1 (mark; ▲) with the solid nozzle. We can not see any difference in the field gradient in both cases. In other words, our present field limitation is not due to particulate matter.

On the other hand, at CEBAF a very distinct effect of HPR on cavity performance is confirmed [2]. Fig.10 is a result of CEBAF. This cavity was electropolished at KEK by 120 μm and rinsed with our HPR system (with the flat nozzle). Then we sent it to CEBAF under vacuum. The cavity was measured after disassembled in the clean room and rinsed with reagent methanol prior to the test. After this measurement (mark; x), it was rinsed with CEBAF's HPR system, subsequently rinsed with methanol in the clean room, then tested (mark; •). The accelerating field gradient improved to 28 MV/m (Ep=50 MV/m) without field emission loading. It increased to 33

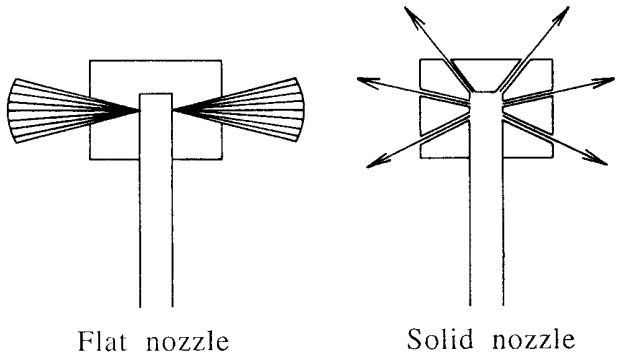


Fig.8 Geometry of nozzles used in the HPR experiment.

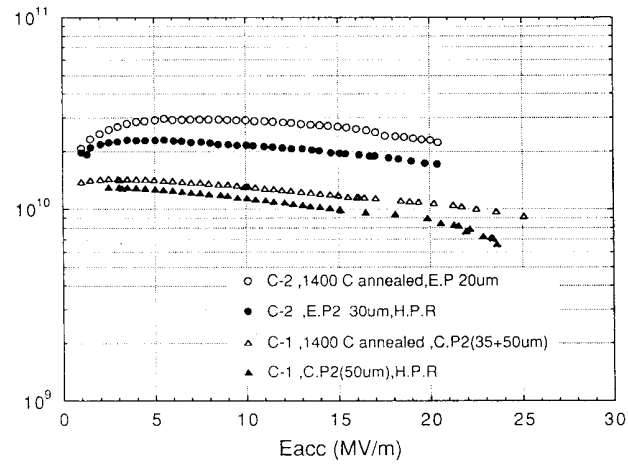


Fig.9 Cavity performance by HPR with at KEK.

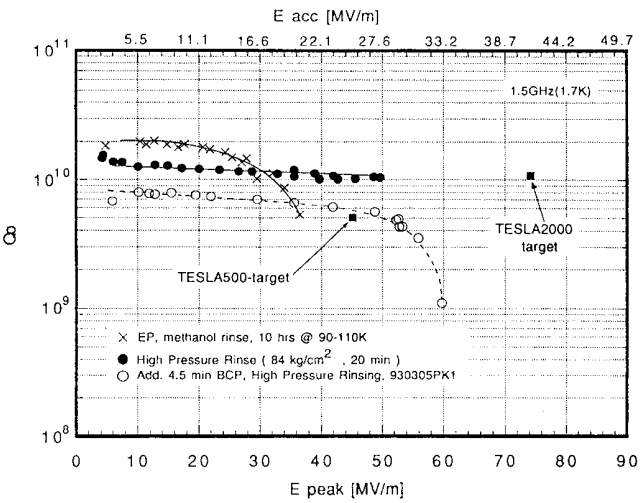


Fig. 10 Cavity performance with HPR at CEBAF.

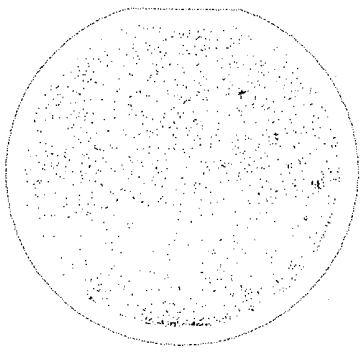
MV/m (Ep=60 MV/m) in the next test with HPR.

Since HPR handles a very high pressurized water, one has to be careful about the contamination from HPR system. It was found later, our flat nozzle was scraped by the high pressure water, and the pressure of water dropped slowly 85 to 65 kg/cm^2 during these HPR experiments. Some cavities suffered field emission loading by this trouble.

E. Effect of megasonic rinsing

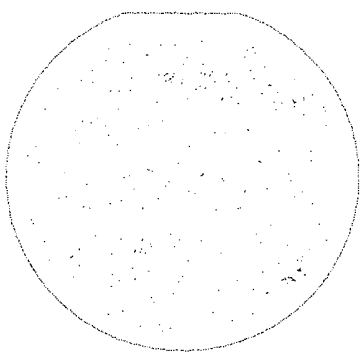
We discussed about megasonic rinsing in the last SRF workshop at DESY but it was just an information from people of semiconductor technology. We evaluated the effect of megasonic rinsing and compared with HPR. We summarize the rinsing conditions and results in Table 1. Fig.11 and 12 are results with HPR and megasonic rinsing in which rinsing conditions correspond to 16 and 19 in Table 1. One can see the powerfulness in megasonic rinsing. Megasonic rinsing enables to eliminate dust particles up to 220; one-fifth of the case of HPR. We have another evidence showing that megasonic is more powerful than HPR. As mentioned before, on the wafer once dried, megasonic rinsing could reduce particles from 10000 to 2650, while HPR to 8500. As a conclusion, we can say megasonic is more powerful tool than HPR to eliminate particle on the surface. We will soon start to test using cavities on megasonic rinsing.

On the other ultrasound agitation, compare the rinsing condition 11, 12 and 15 in Table 1. One can see a tendency that higher frequency is more effective.



Particle size	Count
0.30-1.20 μm	673
1.20-2.01 μm	50
2.01-3.00 μm	305
> 3.00 μm	90
Total	1118

Fig. 11 Residual particles on a wafer surface after HPR; rinsing condition 19 in Table 1.



Particle size	Count
0.30-1.20 μm	158
1.20-2.01 μm	5
2.01-3.00 μm	43
> 3.00 μm	18
Total	224

Fig. 12 Residual particles on a wafer surface after megasonic rinsing; rinsing condition 16 in Table 1.

IV. SUMMARIES

In this paper we discussed about particles from working environment, in chemicals, residual particle on the surface with our standard rinsing procedure, effect of high pressure ultrapure water rinsing, megasonic rinsing, and so on. As a summary, although it has been often said, we can conclude as follows;

1) Do rinsing work under clean environment; in a clean room,
2) Employ powerful methods to eliminate dust particles on surfaces; HPR, megasonic rinsing and so on,
3) Use clean chemicals; EL-G,
4) Don't get dry the surface on the way,
5) Be careful about the rinsing parts taken into cavities; water tubes, seals and nozzles.

Addition to these comments,
6) it will be very important to use hot water to take away chemical residues.

On L-band niobium cavities, current rinsing techniques have overcome field emission loading at least up to 50 MV/m in electric surface peak field (Ep). It corresponds to the TESLA-500 target. We have almost understood how to control particulate contamination. What is the next field limitation on niobium superconducting cavities; thermal magnetic breakdown [2], Japanese Q-disease [6]? Maybe material imperfection will limits the higher field. The history of superconducting cavities will come back again. We will go back to niobium material.

Rinsing techniques developed in superconducting cavities is useful to upgrade general accelerator technologies. We recently started to apply HPR to S-band normal conducting cavities for laser triggered RF-guns [9]. We have achieved an Ep $\geq$ 300 MV/m with low dark currents. The field enhancement factor (ß) was 37. This value is clearly small comparing to that of copper cavities normally prepared, in which ß is 50-60.

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