Bonding of a microwave-absorbing ferrite, TDK IB-004 with copper for the HOM damper of the KEK B-Factory SC cavities

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Abstract

Need for vacuum compatible bonding of ferrite used as microwave absorbers in accelerators is increasing. Bonding of a TDK ferrite IB-004 with copper has been investigated. Brazing, soldering and Hot Isostatic Press(HIP) have been tried. HIP of pre-sintered ferrite powder seems most promising, although further study is necessary.

Introduction

Need for the development of Ultra-High-Vacuum(UHV) compatible bonding of microwave-absorbing ferrites used as Higher Order Mode (HOM) damper has been increasing [1-3]. There are, however, few literatures on this subject. Thus we have been trying some methods which have been used for ceramic-to-metal bonding. There are a few reviews on the bonding techniques of ceramics[4-6]. In this paper, the results of ferrite bonding tests we have performed with brazing, soldering and HIPping will be presented.

General ideas of bonding

Here, we summarize the features of three bonding methods, brazing, soldering and HIPping briefly, assuming that readers know what brazing and soldering are.

Brazing

For excellent brazing good wettability of filler metals is important. Wettability is usually described by the contact angle of the filler metal to the material as shown in Fig. 1. If this contact angle, θ, is less than 90 degrees, the filler metal is said to wet well.

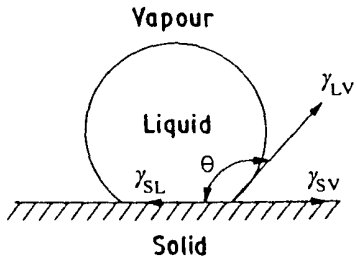

Fig. 1. Schematic representation of contact angle [5].

Wetting of nonreactive metals with ionocovalent oxides were discussed by Li based on the work of adhesion, free energy of formation of the corresponding oxides, bandgap energies of the oxide, which can be translated to the electron density at the interface [7]. The work of adhesion, W, which is defined as the work per unit area to separate a solid/liquid interface is described as

$$W = \gamma_{LV} (1 + \cos \theta) \qquad (1)$$

where γ_{LV} is the surface tension of the liquid and θ the contact angle. As one can seen in eq. (1), work of adhesion increases with the decrease of contact angle and becomes 0 when $\theta = 180°$. This indicates that the bond strength increases with decreasing contact angle, which has been verified experimentally [5].

There are two types of brazing, direct brazing and indirect brazing. In direct brazing, often active component such as titanium is added in the filler metal to improve the wettability since oxides or ceramics have few filler metals that have excellent wettability. In indirect brazing, oxides or ceramics are metallized prior to brazing for better bonding integrity.

Soldering

The biggest advantage of soldering is its low bonding temperature, e.g. normal soldering temperature is around 180°C. This makes the bonding easier and the residual stress that accumulates upon cooling is less than brazing due to the smaller temperature change. The problem in applying soldering to the accelerators will be its outgas and its deterioration with time for long use. We applied ultrasonic soldering for better wetting, which will be described in the later section [8].

Hot Isostatic Press (HIP) [9]

This bonding method uses high pressure and high temperature so that diffusion occurs at the interface. Today, this technique is largely used in Metal-Metal joining, Metal Matrix Composite (MMC), and Ceramic-Metal joining processes. There are two categories for this bonding. One is the bonding of metal or fabricated ceramic with metal. The other is the bonding of powder with metal. The latter has more advantages than the former in that sintering and bonding can be done simultaneously, thereby reducing the process time. With pressurized sintering, the powder can be more densified and strong. In addition, it has more variety or flexibility on the shape.

Bonding tests

Results of the tests of brazing, soldering and HIPping we have performed are described here. All the tests were carried out for bonding of TDK ferrite IB-004 onto copper.

Brazing

As mentioned earlier, there are two types of brazing, direct bonding with a filler metal and indirect bonding with a metallized ferrite with

Direct brazing

Brazing without using pre-coating is called direct brazing. It is simple and straightforward, but not many braze metals wet well with oxides. Typical values of contact angles of pure metals on oxides (Al_2O_3, SiO_2, ZrO_2, MgO, etc.) lie between 80 and 145° [10]. Usually an active metal such as Ti is added to a normal braze metal to improve wetting. The filler metals we have tested are listed in Table 1 with the test conditions and the results. In all the tests, 2cm x 2cm x 4mm ferrite tile was used to avoid cracking from thermal stress [1]. The vacuum during the tests were about $2x10^{-4}$ Torr. After confirming that the absorption characteristics of our ferrite does not change after 870 °C x 40 min. heating at $1x10^{-5}$ Torr, we have been performing the braze tests only in vacuum to prevent oxidation of copper and reduce voids.

Table 1. Results of sample tests with different filler metals

Sample No.	Filler metal	Composition	Braze Temp. (°C)	Braze time. (min.)	Pressure (kg/cm^2)	Braze result
1-7	BCuP5	5P/15Ag/80Cu	670	10	0.007	Bad
4-2	BAg In-Ti 50μm film	59Ag/27.25Cu 12.5In/1.25Ti	850	20	0.096	Bad
4-3	BAg-8-2Ti 50μm film	70.5Ag/27.5Cu/ 2Ti	850	20	0.096	Bad
5-1	BAg-8 100μm film	72Ag/28Cu	830	20	0.096	Fair[1)]
5-2	BAg-8 100μm film	72Ag/28Cu	830	40	0.096	Fair
5-3	BAg-8 100μm film	72Ag/28Cu	850	20	0.096	Fair[2)]
5-4	BAg-8 100μm film	72Ag/28Cu	850	40	0.096	Fair[3)]
5-5	BAg-8 100μm film	72Ag/28Cu	870	20	0.096	Fair[3)]
5-6	BAg-8 100μm film	72Ag/28Cu	870	40	0.096	Fair[3)]
6-3	Ti/BAg-8 20/100μm film	Pure Ti + 72Ag/28Cu	920	10	0.096	Bad
6-4	BAg-8/Ti 100/20μm film	72Ag/28Cu + Pure Ti	920	10	0.096	Bad
8-1	BAg-8 100μm film	72Ag/28Cu	850	40	75	Good[4)]
8-2	BAg-8 100μm film	72Ag/28Cu	850	40	25	Good
8-3	BAg-8 100μm film	72Ag/28Cu	850	40	7.5	Good

1) "Fair" means that bonding was possible, but weak with many voids.
2) Delamination in the ferrite near the interface occurred upon cutting.
3) Crack when being cut.
4) "Good" means no visible voids in X-ray test and no breaks when being cut.

The ramping rate was 13°C/min. and cooling rate was about 6°C/min. for all the tests. Before the tests, the ferrite surface was finished with #800 paper and degreased with acetone by wiping the surface with a cloth.

When using active filler metals that contain Ti, a brittle material was formed at the interface, resulting in all the unsuccessful tests. Finally, we decided to use BAg-8 and started optimizing the braze conditions as samples 5-1 through 5-6 in Table 2. Even though we could not get an excellent bonding, the optimum temperature and time was 850 °C x 40 min. Some ferrite, however, cracked when being cut. Getting a hint from the HIPping test which will be shown later, we applied more pressure hoping for better strength as samples 8-1 through 8-3 in Table 2. The results showed better strength. All 3 samples did not break upon cutting. The X-ray test also showed no voids as shown in Fig. 3, where the sample without pressure is also shown. Figure 2 shows the X-ray test setup. The detectable void thickness is less than 50μm and the voids shown in Fig. 3 are 300-500μm thick. X-ray test was once compared with ultrasonic imaging and showed excellent agreement [11].

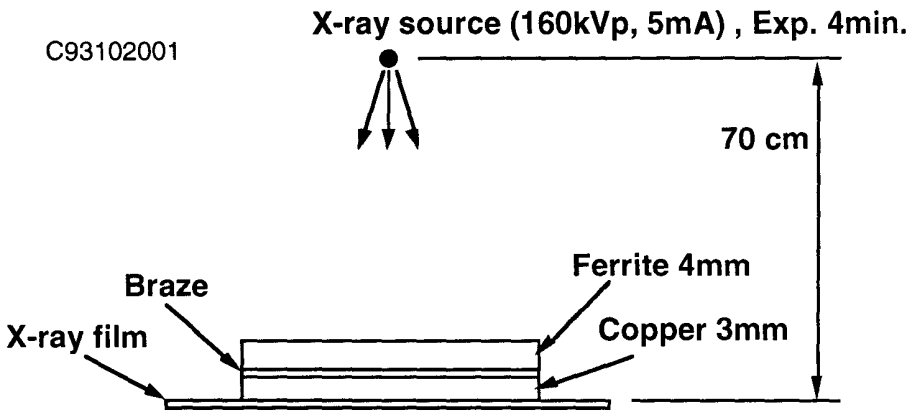

Fig. 2. Schematic representation of X-ray imaging setup.

Fig. 3. X-ray images of the brazed sample with and without pressure.

The problem in applying a high pressure from ferrite side is that, although there is no cracks at the braze interface, there appear cracks on the opposite surface of the ferrite presumably due to the applying pressure and thermal stress upon cooling, which is also attributed to the low tensile strength of the IB-004, 4.4MPa. The optimization of the pressure is under way now.

Indirect brazing

To help the ferrite wet well against the filler metal, we tried to coat the ferrite with three layers of metals, i.e. from ferrite side Ti , Pt and Au of 0.2 μm thick each. This coating is normally used to bond SiC to metals at Hitachi Co., Ltd. The coating itself was firm and excellent, but the result of the brazing tests using both BAg-8 and BCuP were unsuccessful probably due to the inclusion of Ti in the coating because the interface looked similar to the direct braze with the Ti-contained filler metal. We stopped going further with this approach since this method is more complex and expensive compared to the direct brazing.

Soldering

Ultrasonic soldering was tested to improve the wetting against ferrite. We have tried a solder named Cerasolzer manufactured by Asahi Glass Company,

which was developed for soldering glass and ceramics with metals. This solder is similar to the usual Pb-Sn solder except for a little addition of Zn, Sb, Al, Ti, Si and Cu. Table 2 shows the properties of Cerasolzer W123 and W297 which we have used. Due to the easiness and relatively low temperature needed, this soldering has been used for the high power tests of ferrite samples in air [12].

After putting the solder on both ferrite and copper with the vibrating bonder, we put them together and held at slightly higher temperature than the melting point.

Figure 4 shows the X-ray image of the soldering of 6 cm x 6cm x 4mm ferrite tile with 3 mm-thick copper. No typical voids, i.e. round bubbles, were seen, but string-like lines were seen. When the sample was cut, however, it split in the interface. The micrograph of the cracked cross section is shown in Fig. 5. As one can see, cracks run not at the interface but in the solder. This might have been caused by the oxidized surface of solder during brazing because we performed the soldering in air. We are planning to do the next test in either Ar or N_2 atmosphere.

Table 2. Solders used for ultrasonic soldering, Cerasolzer.

Name	Composition	Melt. Point (°C)	Hardness 20°C, H_V	Th. Exp. Coeff. 15-110°C ($x10^{-6}$)	Elec. Resist. ($x10^{-6}\Omega$ cm)
W123	50Cd/46In + Zn, Sb	123	4.63	-	10.4
W297	90Pb/5Sn + Al,Sb,Zn.Ti,Si,Cu	297	12.6	28.7	21.0

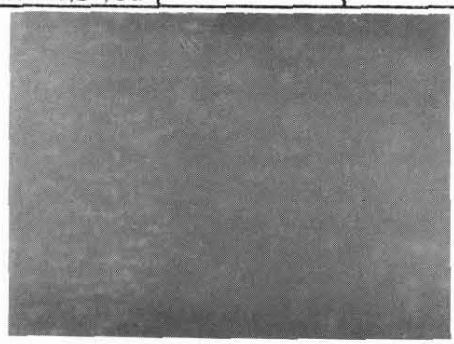

Fig. 4. X-ray image of the ultrasonically soldered joint

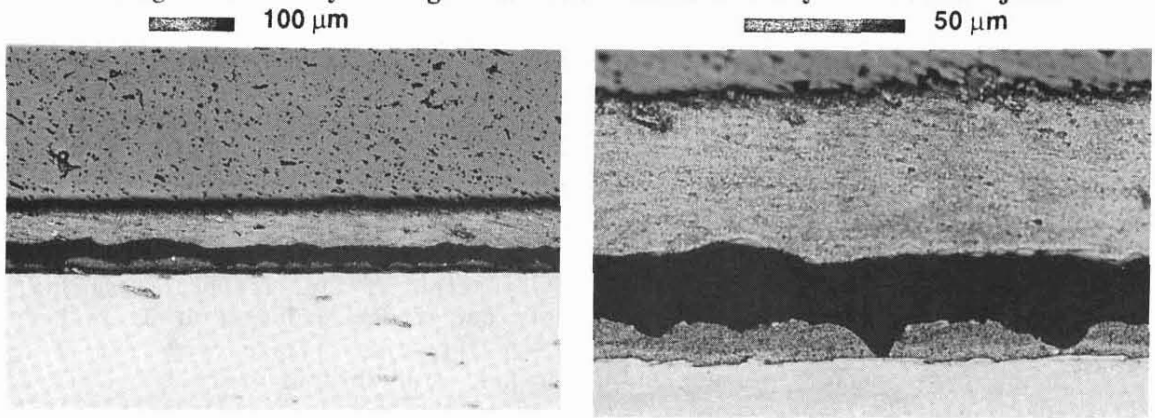

Fig. 5. Micrograph of the cracked interface of soldered joint, W297.

The outgas from the 6cm x 6cm x 4mm ferrite and 8cm x 8cm x 3mm copper soldered together with Cerasolzer W297 was measured to be $2.5x10^{-7}$ Torr l/sec at 25°C after 163 hours of baking at 160°C. The outgassing rate at 160°C was $5.2x10^{-7}$

Torr l/s. Compared to the outgassing rate from HIP bonding, 2.5×10^{-9} Torr l/s at 22°C after 25 hours of baking at 140°C [12], this outgas is 2 orders of magnitude higher and it will be intolerable in the accelerator. Therefore, we are also looking for low outgas solders.

HIPping

As briefly mentioned above, there are two approaches in HIPping. One is to press the fabricated ferrite tiles on the copper surface and the other is to press pre-sintered ferrite powder so that bonding and sintering occur simultaneously. They are labeled as tile HIPping and powder HIPping, respectively, below.

Tile HIPping

To know if the standard IB-004 tiles can be bonded by HIPping, we tried to HIP 6cm x 6cm x 4mm ferrite tiles with 6cm x 6cm x 1cm OFC plates using a 304 stainless steel capsule. The configuration of the canning is shown in Fig. 6. We tested various insert metals such as Ag, Ni and Ti as well as no insertion. Either Boron Nitride (BN) or zirconia powder called zircon flour was used to prevent the stainless steel from bonding with copper and ferrite. Before HIPping, the OFC was degassed at 900°C for about 1 hour. Also, after set up in the capsule, it was evacuated up to 1×10^{-5} Torr. Figure 7 shows the temperature and pressure pattern operated.

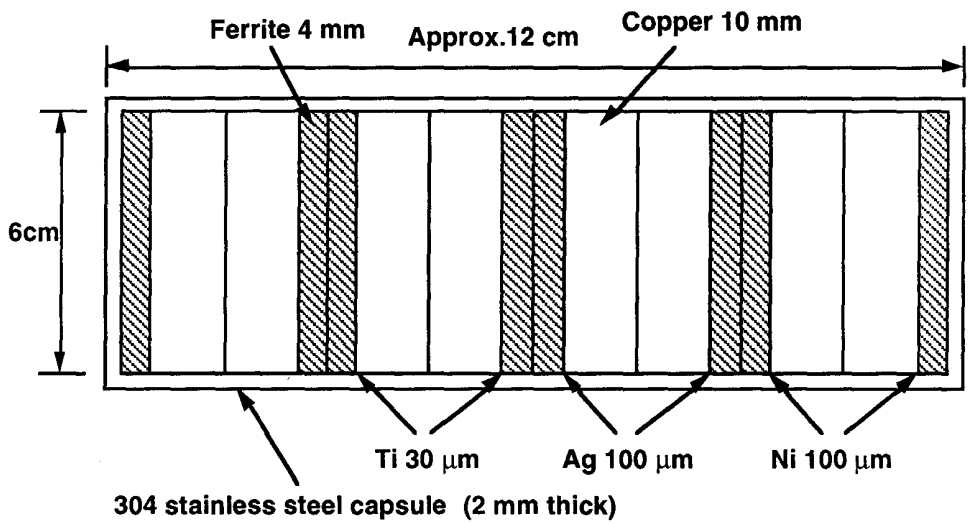

Fig. 6. Configuration of the HIP sample test

The result showed excellent bondings except Ti. Brittle material was formed and delaminated at the interface in the case of Ti. For others, there were no cracks upon cutting. After the HIP test, we cut the sample for X-ray test, outgas test, thermal conductivity test, ε, μ measurement and electric conductivity measurement. Figures 8 and 9 show the X-ray test and the micrograph of the cross section. As one can see in these figures, there are no voids and the copper fused into the surface asperities completely.

Figures 10 through 12 show the SEM images of the bonding interface and the Energy Dispersion X-ray (EDX) analyses of the corresponding metals. Dense white dots show the existence of the corresponding metal, although equally spaced low density dots are noise.

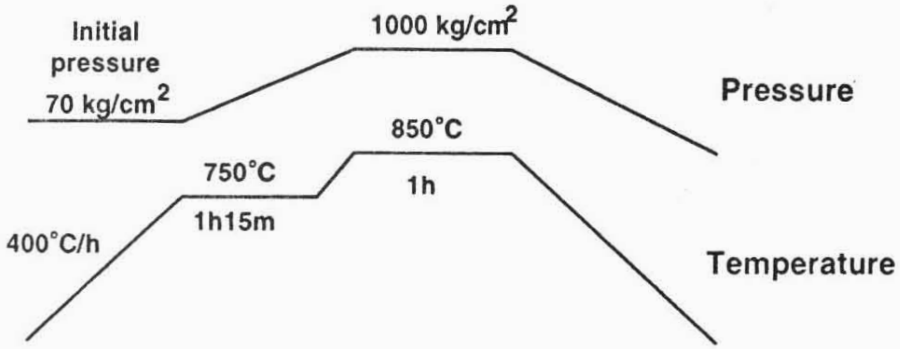

Fig. 7. Temperature and pressure pattern in Tile HIPping.

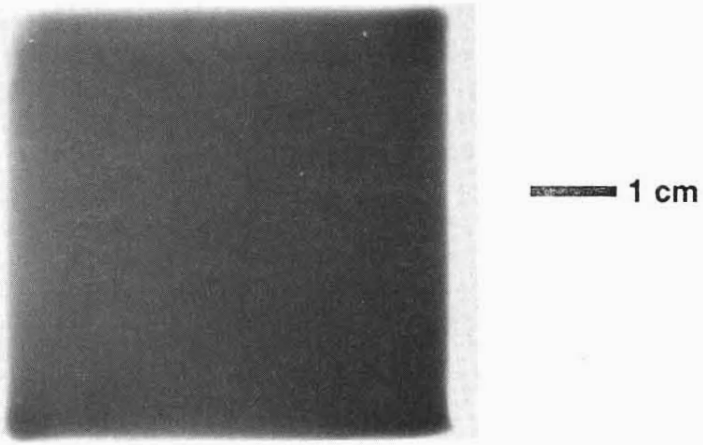

Fig. 8. X-ray image of the HIPped ferrite tile on copper (6cm x 6cm). No insert metal.

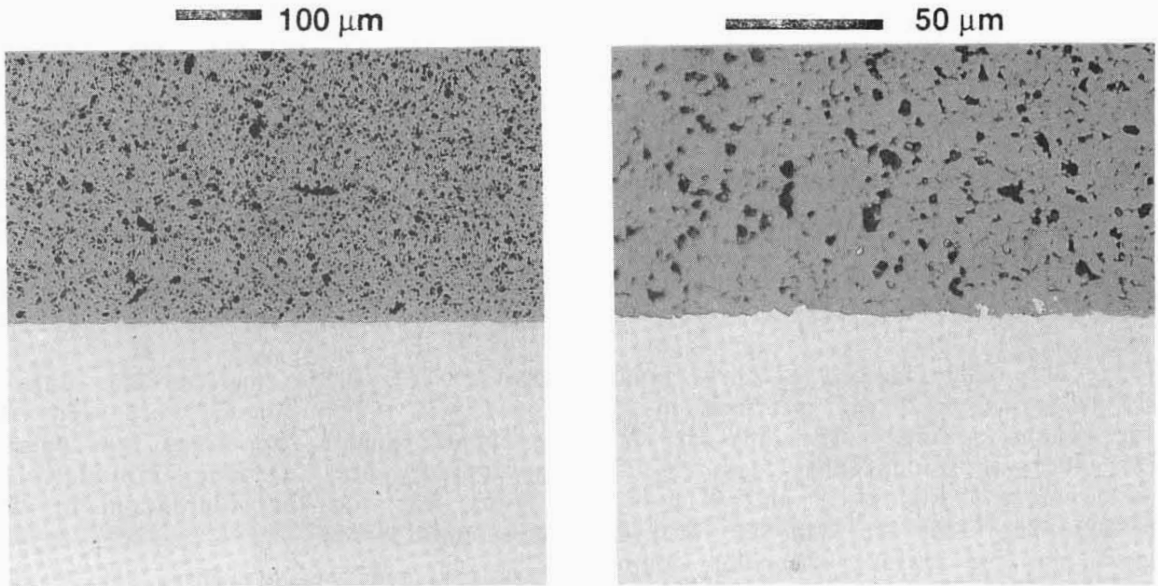

Fig. 9. Micrograph of the interface of HIPped ferrite on copper. No insert metal.

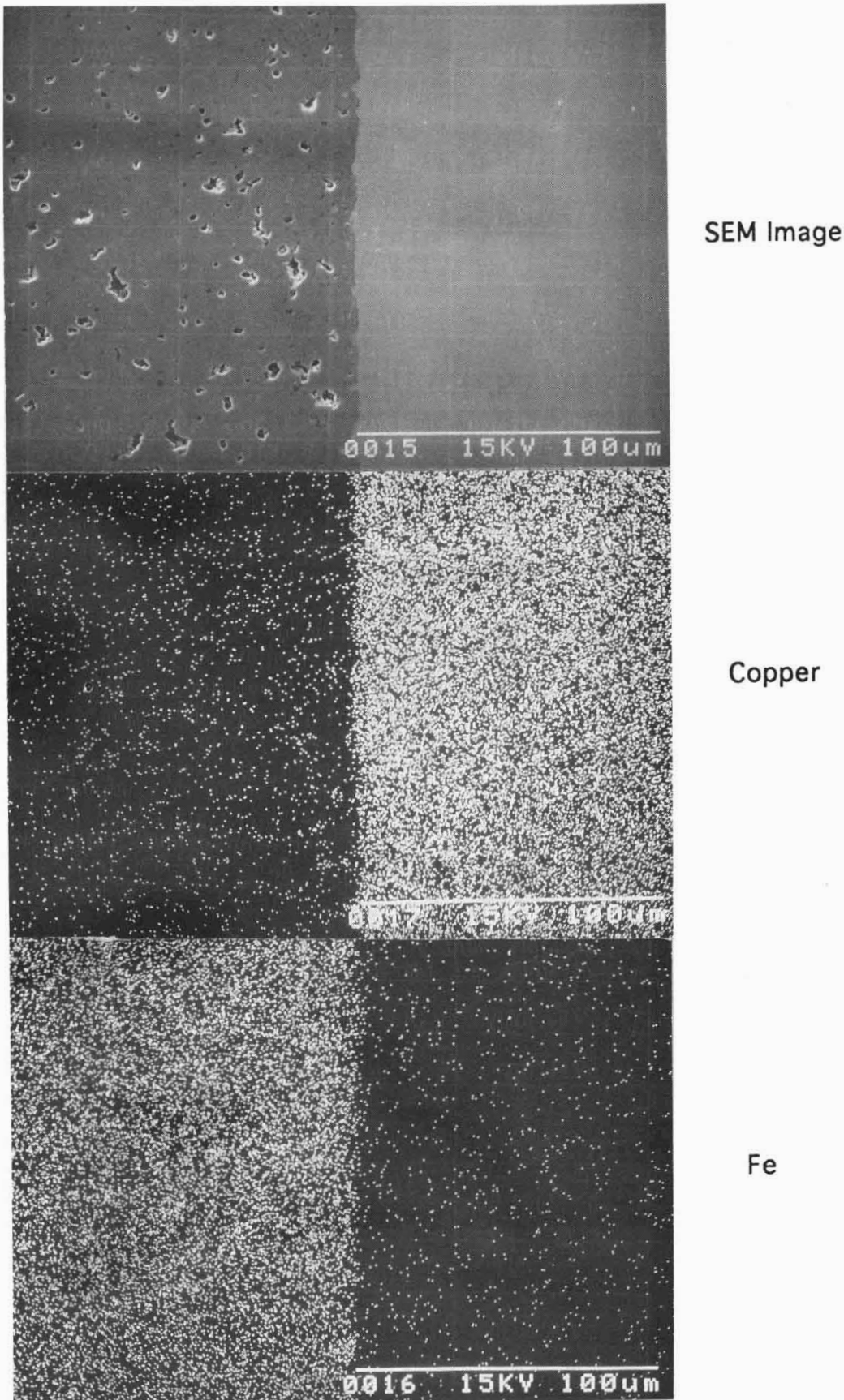

Fig. 10. SEM image and EDX analysis of the bonding interface. No insert metal.

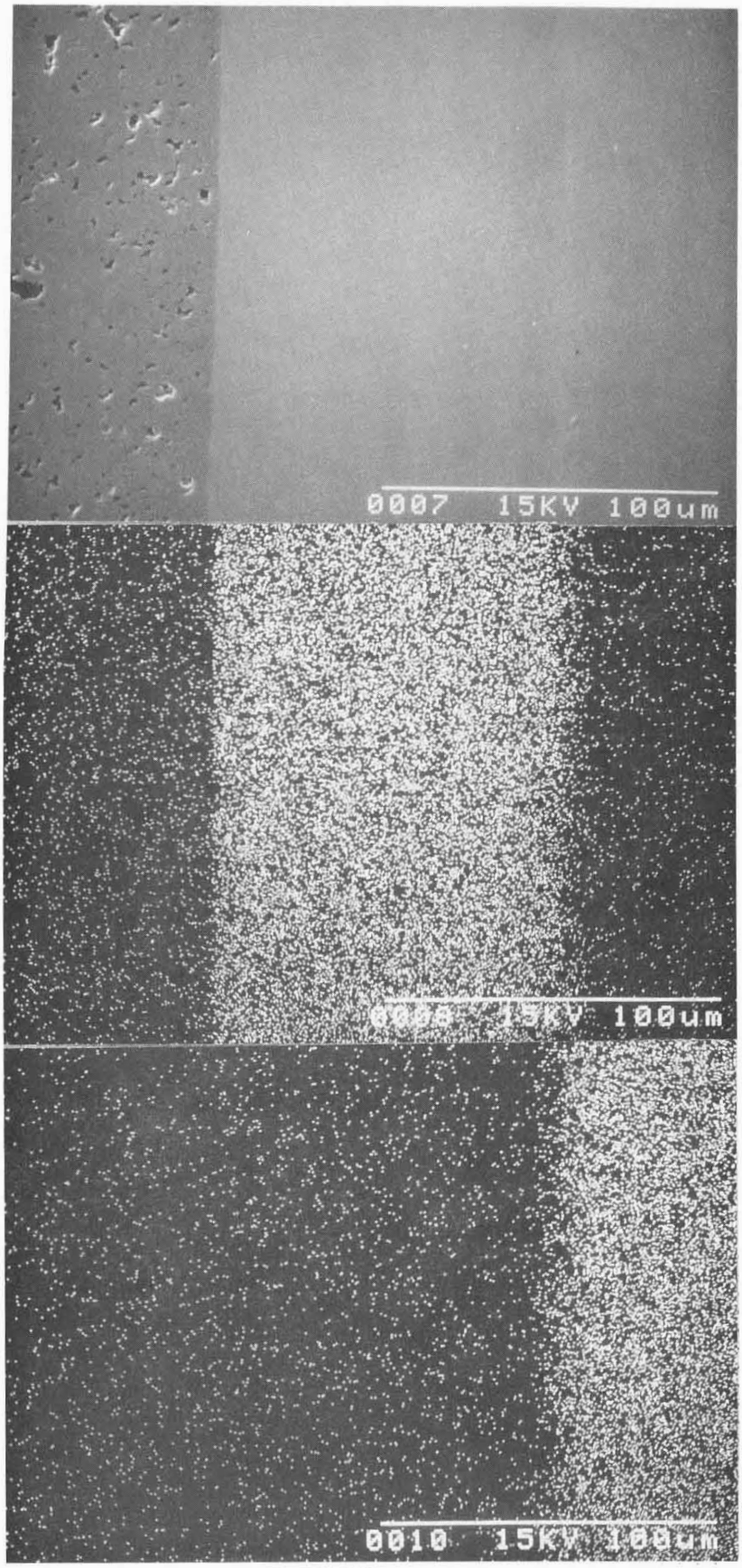

Fig. 11. SEM image and EDX analysis of the bonding interface. 100μm - thick Ni as insert metal.

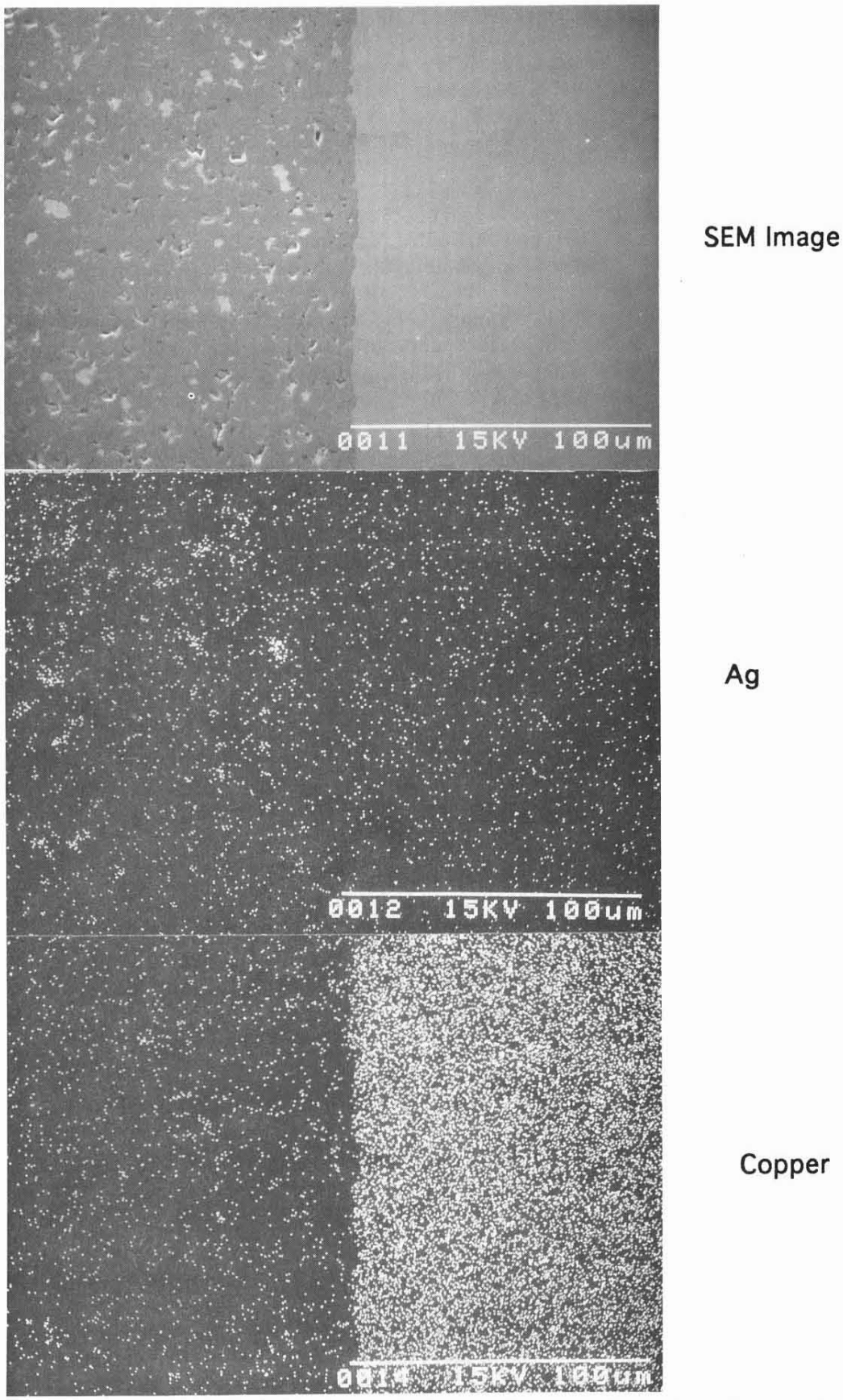

Fig. 12. SEM image and EDX analysis of the bonding interface. 100μm - thick Ag as insert metal.

As one can see in these figures, all the bondings look firm and there is no void at the interface, although there are pores in the ferrite. The silver layer disappeared probably because the HIP temperature exceeded the eutectic point of silver and copper, 780°C. There seems little diffusion between ferrite and metal, although there is some diffusion between Ni and copper. Silver was detected in the pores of the ferrite as shown in Fig. 11, which is an evidence of the melting of silver.

Powder HIPping

Before trying to HIP powder onto the inner surface of the copper cylinder, to check how electromagnetic properties changes in sintering under high pressure, we tried to hot-press the pre-sintered powder uniaxially with carbon press at 1000°C x 2h and 300 kg/cm^2, using a small sample in vacuum. Figures 13 and 14 show the schematic setup and the temperature pattern. The applied pressure, 300kg/cm^2, was same from the beginning through the end. This pressure was limited by the break of the carbon holder.

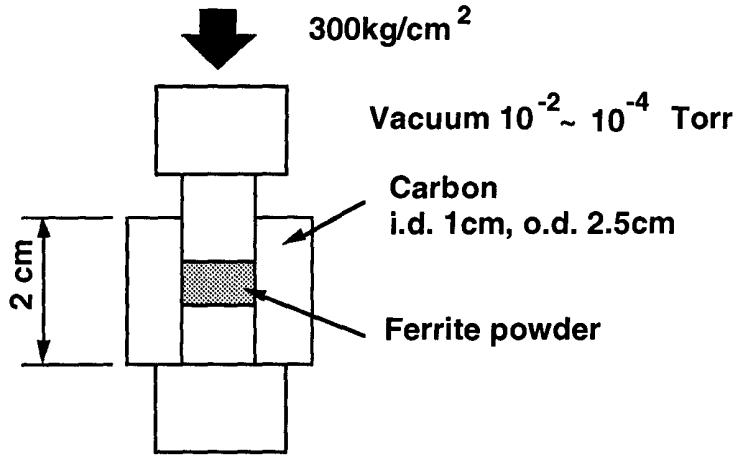

Fig. 13. Setup for hot-press test of ferrite powder.

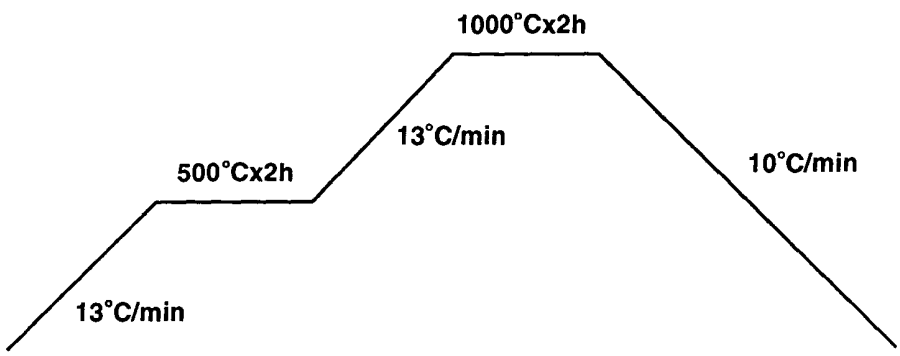

Fig. 14. Temperature pattern in Hot-pressing ferrite powder. Pressure was same during the operation, 300kg/cm^2.

Figure 15 shows the micrograph of the hot-pressed ferrite in comparison with the standard IB-004 which is sintered in air at 1 atm. As one can see in the

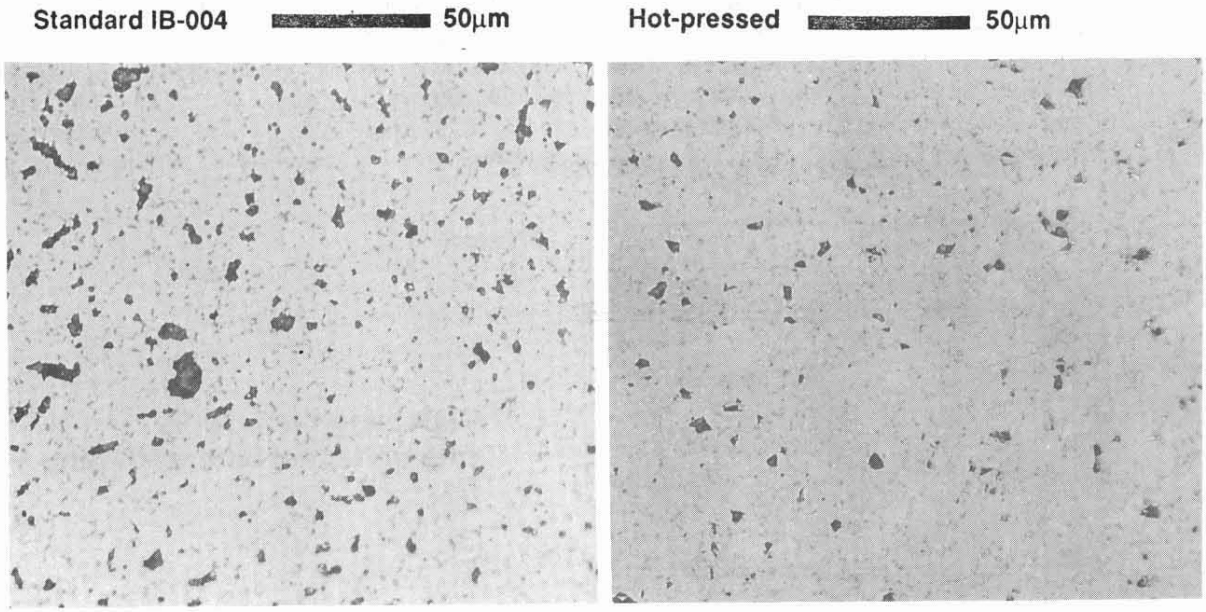

Fig. 15. Micrograph of hot-pressed ferrite in comparison with standard IB-004.

figure, there is no large pore and the number of the pores decreased. Magnetic loss did not change much at the frequencies of interest, although at lower frequencies it decreased significantly [12]. Other measurements showed that the density increased by 8 %, the porosity decreased from 11% to 4 % and the thermal conductivity increased by 18 % as shown in Table 3. Although we have not measured the outgassing rate, we expect it to be lower than the standard one because of the lower porosity. With these encouraging results, we started investigating on the process to fabricate a ferrite layer on a large copper tube by HIPping.

Figure 16 schematically shows the process through which we are currently planning to fabricate HOM loads. As a first step, we tried to HIP the powder on the inner surface of a 5 mm-thick copper cylinder having an inner diameter of 109mm, with which 304 stainless steel are also tried to be HIPped at both ends to make it easier to weld end flanges later. This is about 1/3 diameter of the full-scale load for the SC B-factory cavity. The purposes of making this small load are 1) to check the viability of the above HIPping process and 2) to test the power handling capability of this type load using available 2.45GHz power source.

In addition to this cylinder, we tried to HIP two small samples for property measurements such as electromagnetic loss, mechanical strength of bonding, etc. The shape of this sample after capsuling is shown in Fig. 17, in which ferrite powder was sandwiched with two 6 cm-diam. and 1cm thick copper disks.

The HIP procedure we took was 1) degas the OFC at 870°C, 2) assemble the can and TIG weld the lid of one end, 3) fill the can with ferrite powder, 4) weld another lid and a port for evacuation, 5) evacuate the can and degas the powder at 300°C for a few hours until the pressure goes down to $1x10^{-5}$ Torr, and 6) chip off the can and HIP with Ar. In this first HIP test, powder packing was done by hand with a stainless steel rod and the packing density of the filled powder was only about 40 % of the hot-pressed ferrite. For HIPping powder the desirable packing density is more than 60 %. The powder size, ~3μm, seems too small for a good HIP. However, considering the costly and time-consuming process we should add to get desirable size powder, we decided to continue using this size powder.

The temperature and pressure pattern of HIP is shown in Fig. 18. Taking into account the melting temperature of copper, 1080°C, we decided the HIPping

temperature to be 850°C, which is 150°C lower than that in the powder hot-pressing mentioned above. We also decided to hold the temperature at 300°C for 2 hours in the course of cooling to reduce the residual stress in the copper, thereby reducing the residual compressive stress in the ferrite.

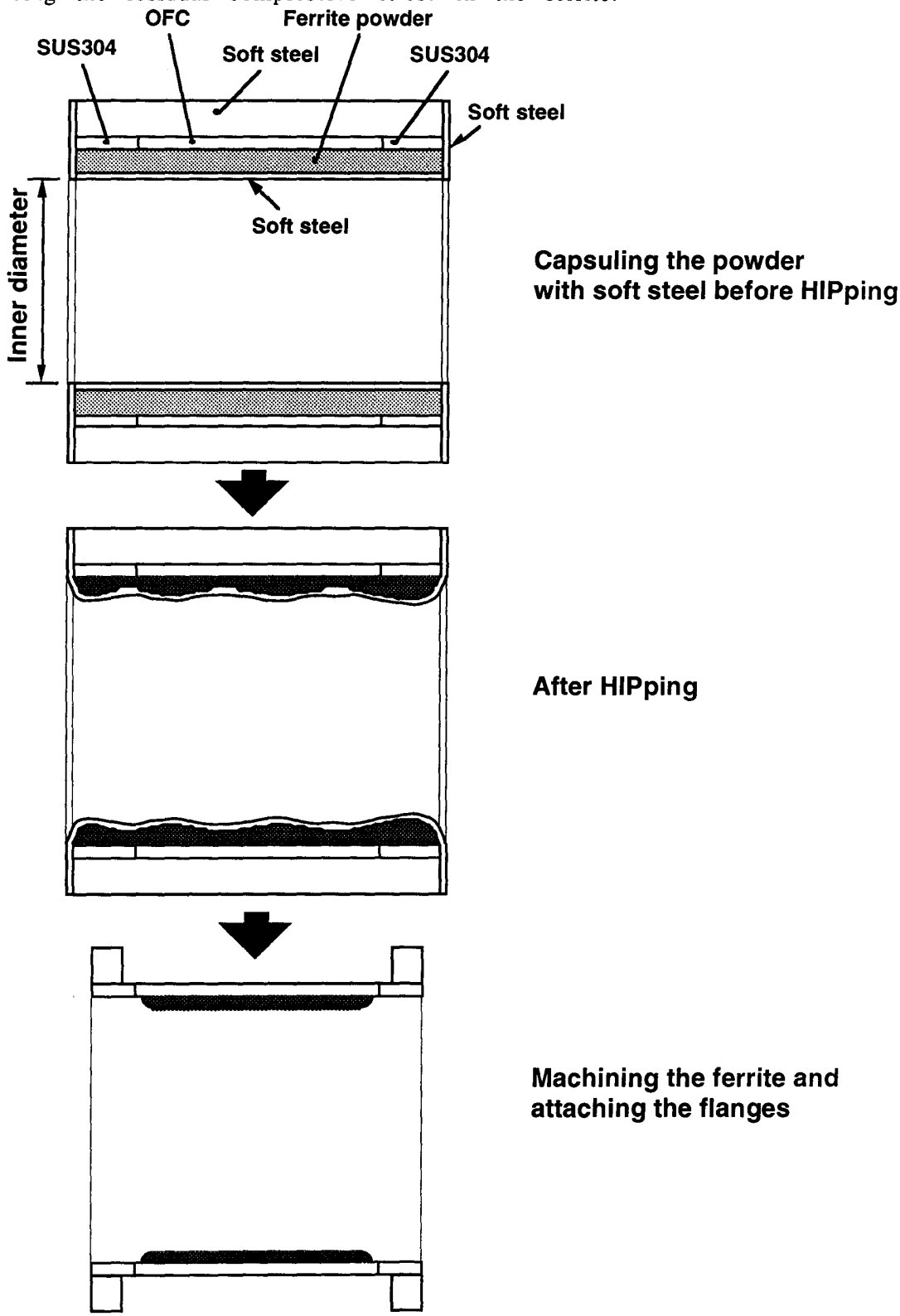

Fig. 16. Plan of the fabrication process of HOM load by HIPping.

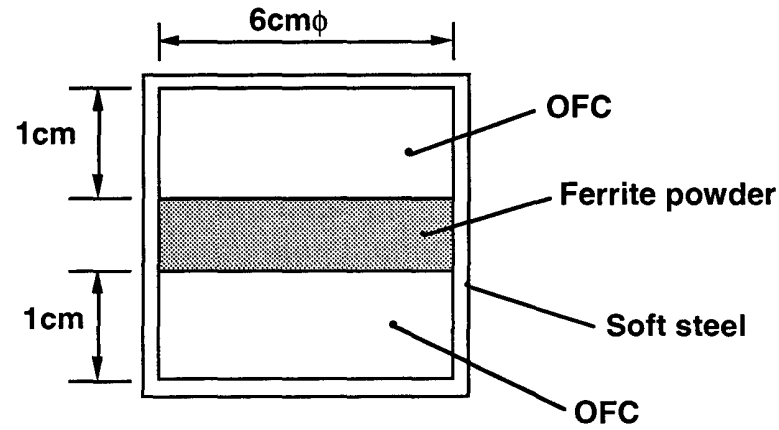

Fig. 17. Capsuling of a HIP sample for property tests.

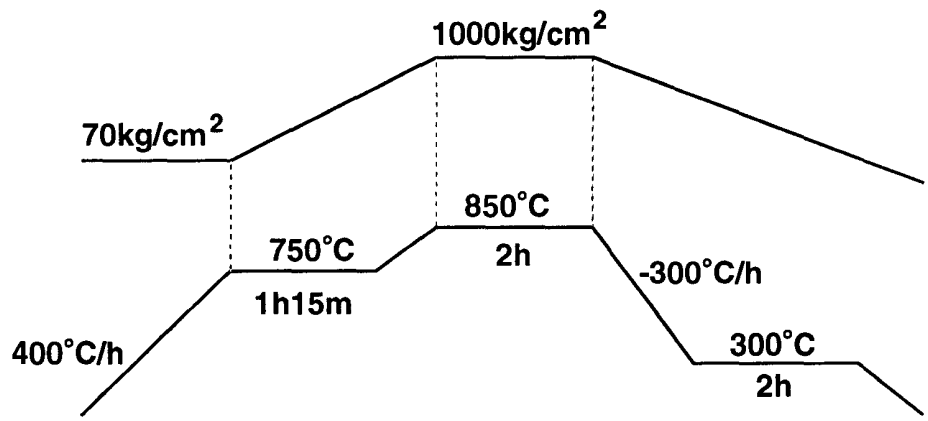

Fig. 18. Temperature and pressure pattern of the 1st powder HIP.

The result of this 1st powder HIP test was unsuccessful. The contraction of the can, 0.9mm, was unexpectedly small compared to the predicted contraction of about 6mm. The possible explanation of this is a small leak during the pressing. Once a small leak occurs, the Ar gas enters the can and the radial pressure disappears. However, the small sample shrank as we expected and we could check some properties, although we could not get the bonding of copper and ferrite due to too much radial shrinkage of the can which surrounded the ferrite powder leading to a crack at the interface. We measured the RF properties and density. The RF loss decreased but still usable, but as one can see in Table 3, the density did not increase much, compared to that of hot-pressed sample, which implies that the sintering was not complete and densification had not occurred completely.

In the 2nd test, we changed the way to weld the steel lid since, we think, the leak occurred at the weld seam. We also changed the holding temperature from 850 °C to 900°C and its pattern as well so that the sintering will be completed and enough densification will be obtained. As a result, the contraction of the can occurred as we expected and we confirmed that a certain ferrite layer had been formed when we took off the inner steel wall with a lathe. It is under final machining of the ferrite now. The properties of the other sample will be checked soon, which will be reported elsewhere.

Table. 3. Summary of IB-004 formed in various ways.

Type	Density (g/cm^3)	Porosity (%)	Th. Cond. (W/mK)	Outgass[1] (Torr l/s cm^2)
Standard[2]	4.84	11	6.28	$5x10^{-11}$>
Tile HIP 850°Cx1h, 1000kg/cm^2	4.95	9	6.49	$1.4\text{-}2.5x10^{-11}$
Powder Hot press 1000°Cx1h, 300kg/cm^2	5.23	4	7.39	-
Powder HIP 1st 850°Cx2h, 1000kg/cm^2	4.95	9	-	-

1) At room temperature after 140°C x 25h bake.
2) Sintered in air at 1 atm.

Conclusion

Bonding of microwave-absorbing ferrite, TDK IB-004, has been studied for the application to the Higher-Order-Mode damper in the accelerator, especially for the B-Factory superconducting cavities. We have been investigating three approaches, 1) vacuum brazing, 2) ultrasonic soldering and 3) HIPping. For vacuum brazing, the best bonding could be obtained using BAg-8 filler metal plus additional pressure between ferrite and metal. For soldering, current problem is the cracking in the solder, although there is no void, and the excessive outgass. We are planning to change the soldering material and atmosphere from air to Ar or N_2 because the cracking is thought to be due to the oxidation of the solder.

Finally for the HIPping, we tried HIPping tiles and powder. The X-ray test and micrograph showed excellent bonding. The powder HIPping seems very attractive and promising from the 1/3 scale model tests. Hence it is currently under intensive study.

Acknowledgments

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