Performance of superconducting microwave devices passivated with dielectric materials

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We present a set of experiments which show that three dielectric processing variables in particular affect the performance of superconducting microwave devices: processing time and temperature, moisture content of the dielectric material, and surface interactions with the high temperature superconductor (HTS). The changes in microwave performance of a straight-line microstrip resonator before and after passivation were quantified by measurements of the loaded and unloaded quality factors for each resonator. Dielectric materials of varying moisture content were used. The dielectrics were processed at different times and temperatures. This study shows that the degradation of the microwave devices can be minimized by choosing dielectrics which (i) have a low moisture content, (ii) interact as little as possible with the HTS surface, and (iii) can be rapidly processed at relatively low temperatures. © *1997 American Institute of Physics.* [S0003-6951(97)02337-1]

It is widely recognized that high temperature superconductor (HTS) circuits offer dramatic performance improvements over conventional technologies for a wide variety of circuit functions. This is particularly apparent in the microwave electronics arena where HTS microwave filters, delay lines, antennas, and oscillators show orders of magnitude better performance than conventional circuitry.[1] In addition, HTS circuits can be significantly smaller in size than other devices. As HTS technology moves toward high volume production, increased emphasis is being placed on reproducibility, reliability, and cost. An integral part of this transformation is the development of passivation layers which protect the HTS structures from physical and electrical degradation. The environmental stability of HTS is a fundamental concern for many device applications, for example, $YBa_2Cu_3O_{7-x}$ compounds and a variety of intermediate species.[2–6] Several approaches to passivation have previously been studied, including incorporating Ag in the film and formation of native compounds.[7–9] It was found that vacuum exposure resulted in little increase in contact resistance, indicating no surface oxygen loss.[10] Other attempts to passivate HTS include substitution of the cation in the HTS, and the application of diamond like carbon films.[11,12]

The successful dielectric layer will improve reliability without sacrificing performance and will protect circuits during testing, assembly, and in-service usage. To realize a stable, dielectric passivating material, it is particularly important that the processing conditions required to apply and cure the dielectric material be compatible with HTS surface chemistry. HTS is sensitive to high temperature processing, especially in moist or reducing atmospheres.[2–9] Traditional passivation processes cannot be used without sacrificing some level of performance.

In this letter, we present the results from a study where the change in microwave performance was measured as a function of the dielectric material and its processing conditions. The study shows that the HTS degradation is very sensitive to the dielectric processing time and temperature, and the specific properties of the dielectric material (e.g., moisture content and surface interactions). It is also shown that HTS degradation due to ion milling damage during patterning can be reversed by the use of the proper dielectric material and cure conditions.

Double sided $Tl_2Ba_2Ca_1Cu_2O_8$ (TBCCO) films were prepared by a two step process. An amorphous film was deposited by pulsed laser deposition onto a two inch diameter substrate at room temperature. The samples were then crystallized into an epitaxial $Tl_2Ba_2Ca_1Cu_2O_8$ film by annealing at 820 °C for several hours. The resultant films were then patterned using standard lithographic procedures into devices which were diced and mounted into test fixtures. The test vehicle used to quantify the changes in microwave performance before and after passivation was a straight line microstrip resonator 7 mm long×150 μm wide. The loaded quality factor was measured for each resonator. The unloaded quality factor is calculated from

$$1/Q_0 = 1/2Q_{\text{coup.}} + 1/Q_{\text{diel.}} + 1/Q_{\text{cond.}} + 1/Q_{\text{rad.}}, \qquad (1)$$

where (Q_0, a measure of the total loss of the circuit) $Q_{\text{coup.}}$ is the coupling Q, $Q_{\text{diel.}}$ is the dielectric Q, Q_{cond} is the conductor Q, and $Q_{\text{rad.}}$ is the radiation Q. $Q_{\text{diel.}}$ and $Q_{\text{rad.}}$ are unknown, but because the geometry is the same for each test, it is assumed that relative changes in calculated, unloaded Q are reflections of changes in $Q_{\text{cond.}}$.

The Q of the TBCCO resonators was measured and used to evaluate the enhancement or degradation of the microwave device properties of the thin film superconductor. The passivation materials and processing conditions used in this study are listed in Table I. The processing conditions were selected so as to minimize the time of exposure at high temperatures, yet still produce acceptable mechanical and electrical properties for the dielectric material. For the spin-cast dielectrics, the solutions were dispensed onto the surface of the device in static mode and spun for 30 s at 3000 rotations per minute (RPM). A final polymer thickness of approximately five μm was desired. The polymers were cured in

TABLE I. Processing conditions of the passivation materials used and the average percent change in Q observed.

Passivation material	Processing conditions	Average % age change
TEFLON AF 1601 (6%)	165 °C 5 min/330 °C 12 min	9
TEFLON AF 1601 (6%)	165 °C 5 min/250 °C 12 min	25
Cyclotene 3022 (39% cure)	200 °C for 6 min	4
Cyclotene 3022 (50% cure) w/ adhesion promotor	220 °C for 6 min	−8
Cyclotene 3022 (90% cure)	240 °C for 6 min	−4
Cyclotene 3022 (90% cure) w/ adhesion promotor	240 °C for 6 min	−25
Cyclotene 3022 (90% cure) w/ adhesion promotor	275 °C for 36 s	−22
ULTRADEL 7501	200 °C for 90 min	−20
PROBIMIDE 293	200 °C for 90 min	−35
PI-2540	200 °C for 90 min	−36
ACCUFLO 913EL (SOG)	200 °C for 2 min	−40
AVATREL 9610	220 °C for 6 min	−32
AVATREL 9610	165 °C 5 min/250 °C 60 min	−80
PI-2611	200 °C for 90 min	−51
SiO_2	PECVD 0.5 μm/250 °C	−22
SiO_2	PECVD 0.5 μm/200 °C	−59
Si_3N_4	PECVD 0.5 μm/250 °C	−13

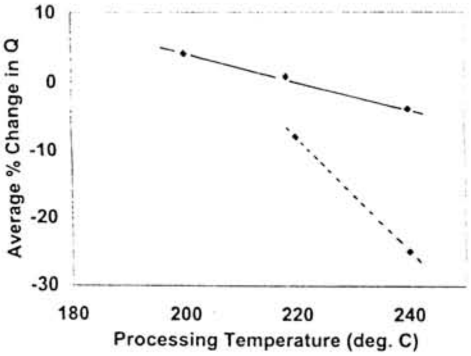

FIG. 1. Effect of the processing temperature of cyclotene on the extent of degradation of the superconducting Q factor. The cyclotene was cured on a hot plate for 6 min under a nitrogen blanket in each case. The dashed line is with the use of an adhesion promoter and the solid line is without the use of an adhesion promoter.

either a nitrogen filled furnace or on a hot plate under a nitrogen blanket. The SiO_2 and Si_3N_4 were deposited by plasma enhanced chemical vapor deposition (PECVD).

The Q for each of the resonators was measured before and after processing of the dielectric material. The average percent change in Q is shown in Table I. Each experiment was performed several times (between two and six repetitions, as given below). The range and average Q for each group of samples prior to processing is given in the text. Positive values indicate that the microwave device properties of the superconductor improved during the processing of the dielectric, while negative values indicate that the superconductor degraded as a result of the dielectric processing. It is known that the superconducting properties of YBCO and TBCCO are very sensitive to its oxygen content which can be affected by the oxygen content of the ambient, particularly at elevated temperatures.[2–10]

The average changes in the Q factors for the resonators were 9% and 25% for Teflon AF 1601 cured at 330 °C (six resonators whose Q ranged from 8329 to 11 544 and average Q was 10 307) and 250 °C (six resonators whose Q ranged from 8439 to 13 730 and average Q was 10 875), respectively. The original degradation in Q (from the as-deposited value) was caused by ion milling damage during resonator fabrication. The mechanism for the improvement in the superconductor properties is not understood (beyond the scope of the present study), but improvements are often tied to changes in the oxygen content of the HTS line which could occur during dielectric processing. The reported residual moisture in Teflon AF is very low, 0.01%, so that additional degradation due to reaction with water is minimized.[13] Although quantitative adhesion measurements were not performed, in comparison to the other dielectrics, Teflon AF 1601 did not adhere as well to the resonators and substrates.

Cyclotene 3022 is a thermoset polymer and also has very low residual moisture, <0.2%. The extent of cross linking can be controlled by time and temperature. Five sets of resonators were fabricated and the results are tabulated in Table I. The lowest temperature process (200 °C for 6 min), 39% cured polymer, gave the most favorable results with a 4% improvement in Q (four resonators whose Q ranged from 9444 to 11 101 and average Q was 10 474). As the temperature was increased to 220 °C (four resonators whose Q ranged from 8649 to 11 570 and average Q was 9714) and 240 °C (four resonators whose Q ranges from 9169 to 17 875 and average Q was 12 643), the average change in Q decreased, as shown in Fig. 1. Of particular interest is the effect of the adhesion promoter. The Q factors were lower when the adhesion promoter was used for processing at 240 (two resonators whose Q ranged from 12 485 to 16 571 and average Q was 14 528) and 275 °C (four resonators whose Q ranged from 11 810 to 14 497 and average Q was 13 120). Thus, the additional chemical bonding to the surface (with adhesion promoter) came at the expense of resonator Q. This supports the observations with Teflon AF, where marginal adhesive forces were correlated with improved Q in all cases. The trade-off between time and temperature for the 90% cured Cyclotene 3022 (240 °C for 6 min vs 275 °C for 36 s) shows that the shorter process time (−22% change in Q vs −25% change in Q) is most likely better. However, the two results are very similar.

Ultradel 7501 (four resonators whose Q ranged from 10 912 to 12 686 and average Q was 11 851), Probimide 293 (two resonators whose Q ranged from 15 757 to 17 335 and average Q was 16 546), PI 2540 (two resonators whose Q ranged from 12 183 to 15 452 and average Q was 13 832), and PI 2611 (six resonators whose Q ranged from 8195 to 15 448 and average Q was 12 339) were each processed at 200 °C for 90 min. The temperature is considerably below that recommended for curing; however, higher temperatures caused considerable degradation of the HTS. The furnace curing is needed because volatile solvents and/or reaction products are released and evolved during the process. The average change in the resonators was negative for all four materials, as shown in Table I. The qualitative observation that exposure of TBCCO to even modestly moisture environments is undesirable for long periods of time is substantiated with these materials.

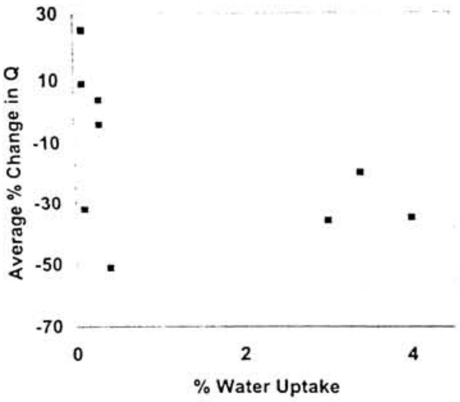

FIG. 2. Effect of the moisture content of the passivation layer on the extent of degradation of the superconductor Q.

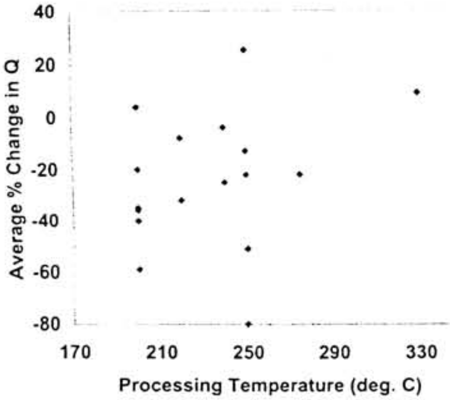

FIG. 3. Effect of the processing temperature for all of the passivation materials on the degradation of the superconductor Q.

Avatrel dielectric polymer is based on polynorbornene and is known for its excellent adhesion. The 220 °C samples used six resonators whose range of Q was 9072 to 14 931 and average Q was 11 938. The 165 °C samples used two resonators whose range of Q was 15 491 to 16 524 and average Q was 16 008. Although the moisture content is very low and the material can be rapidly processed, the change in Q appears to be due to the high degree of interaction between the polymer and the superconductor. Accuflo 913EL spin-on-glass also resulted in considerable degradation of the superconducting films, in spite of the low temperature and rapid process (six resonators whose Q ranged from 10 644 to 13 004 and average Q was 11 891).

Last, several PECVD deposited inorganic dielectrics were investigated. 0.5 μm of SiO_2 was deposited on each resonator. The 250 °C samples used four resonators whose range of Q was 9006 to 13 450 and average Q was 11 328. The 200 °C samples used six resonators whose range of Q was 10 796 to 13 830 and average Q was 12 783. The 250 °C samples of Si_3N_4 used two resonators whose range of Q was 12 159 to 13 173 and average Q was 12 666. In each case, the HTS degraded during processing. It should be noted that the deposition chamber was not equipped with a load lock so that the HTS was exposed to air at the deposition temperature during pump down, which could contribute to the degradation.

It is anticipated that the failure mode of the resonators involves a change in the oxygen content of the superconductor in the near-surface region. This can occur by reaction with water, oxygen, the dielectric material, or the chemicals used in the processing (adhesion promoters, solvents, etc.). Thus, minimizing the high temperature exposure of the superconductor is important. The interactions between these factors is not quantitatively known. Figure 2 shows a scatter plot of the average change in Q vs reported moisture content of the dielectric materials. While there is a general trend that lower moisture content is desirable, the processing conditions of the dielectric and surface interactions are clearly important. That is, we found no high-moisture content material to be acceptable, but having low moisture content is not a sufficient condition for preventing degradation.

The scatter plot of average change in Q vs processing temperature, Fig. 3, shows that while temperature is important, it is only one of several factors. Within a specific material, e.g., Cyclotene 3022, one can define a specific relationship between time and temperature.

From this work, it appears that there are three dielectric processing variables which affect the performance of microwave devices: processing time and temperature, moisture content of the dielectric material, and surface interactions (bonding) with the HTS. Thus, for minimum degradation of device performance, the dielectric should have a low moisture content, interact as little as possible with the HTS surface, and be processed for only a short period at relatively low temperatures. Consequently, the ''best'' performing dielectrics will have acceptable adhesion (but not in excess) and will most likely not be fully cured.

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