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SUBJECT: "ULTRA LOW LOSS CERAMIC DIELECTRICS"

To Members of the Lead Industries Association:

Enclosed is a reprint of an article entitled "Ultra Low Loss Ceramic Dielectrics," which appeared in a recent issue of "The Journal of the American Ceramic Society."

This paper was presented by Bunag and Koenig before the Electronics Division of the American Ceramic Society at Asbury Park last fall. As acknowledged on the last page, the work was made possible by the LIA fellowship grant to Rutgers.

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Ultra Low Loss Ceramic Dielectrics

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The object of this research was to develop ultra low loss ceramic dielectrics. The possible causes of the dielectric loss of ceramic bodies were first analyzed theoretically. Experimental bodies, designed on the basis of this analysis, provide for ultra low dielectric loss.

I. Introduction

The trend toward higher frequency electronic equipment has required the concurrent development of ultra low loss ceramic insulation. There has been extensive work under way along these lines during recent years, although much of this work has not been reported in the literature.[1] These earlier data were reviewed before making the present theoretical analysis.

II. Possible Causes of Dielectric Loss in Ceramic Bodies

If an a. c. electric field is applied to a condenser which is made of a certain dielectric and the electric field does no net work when averaged over one cycle, then the dielectric has no dielectric loss. But actual dielectrics have some losses, in which case the applied electric energy is finally converted to heat. Since the electric field does work on the dielectric, the phase angle between the field and electric current should be less than 90°, e.g., 90° − δ, where δ is the "loss angle." Usually sin δ (power factor) or tan δ (dissipation factor) is used to specify the loss character of a dielectric.

Any process converting the stored electric energy of the dielectric into heat energy can be a cause of dielectric loss. Electrical conduction of a dielectric and time lag of the orientation of a permanent dipole (or of an induced electric dipole moment at ultra high frequency) with respect to the phase angle of an applied electric field are common causes of dielectric loss. These losses are of an electrical origin in the macroscopic sense. When a dielectric consists of a mixture of two or more materials, the Maxwell-Wagner type of di-

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[1] Progress Reports, Contracts DA-36-039-sc-64506, -42877, -38189, School of Ceramics, Rutgers, The State University, The United States Army Signal Research Laboratory, Fort Monmouth, New Jersey.

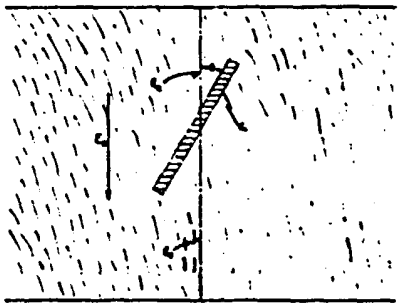

Fig. 1. Mechanical interaction between phases. ϵ_1 = dielectric constant of long cylinder, ϵ_2 = dielectric constant of homogeneous media and E_0 = applied electric field.

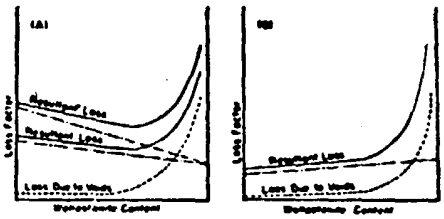

Fig. 2. Assumed diagram of dielectric losses of wollastonite bodies as a function of wollastonite content.

electric loss appears. This loss is due to an accumulation of electric charges on the grain boundaries within the mixture.

Usually, the theoretical dielectric constant and power factor of a heterogeneous substance (a mixture of two or more phases, like a ceramic body) are given in terms of the dielectric properties of the component phases. Several theories have been developed regarding this, such as Weiner and Lichtenecker's formula,[1] Bruggeman's theory,[2] and Nicsel's theory.[3] None of these theories has taken into account the loss due to the mechanical interaction between phases. For an ultra low loss dielectric, there is a possibility that this becomes an important part of the mechanism.

For example, consider a ceramic body containing piezoelectric material such as crystalline quartz. When an electric field E with a high frequency ω is applied to a ceramic, each piezoelectric crystallite in it will tend to oscillate with the same frequency. The amplitude would be very, very small, since the crystallites are surrounded by a glass phase. Due to this oscillation, sound waves of frequency ω will be generated at each crystallite in the ceramic body. The dissipation of the sound wave energy depending on the elastic properties of the ceramic will cause a dielectric loss.

Even though a ceramic does not contain any piezoelectric material, the same kind of mechanical loss may occur. A simple illustration of this is given in Fig. 1, in which a dielectric contains a long cylinder with dielectric constant ϵ_1 embedded in a homogeneous substance of dielectric constant ϵ_2. If an electric field, E_0, is applied, there will be a force (torque) on the rod. If $\epsilon_1 > \epsilon_2$, then the torque will have a tendency to decrease θ. If $\epsilon_1 < \epsilon_2$, the reverse will occur. Thus the rod will tend to make rotational vibrations with a frequency of 2ω, generating a sound wave.

If the flux used for making a ceramic body is not sufficient, numerous voids will be found in the fired body. Since some of the raw materials used in a ceramic body are gas forming materials, gases will be produced when the body is fired. Therefore, even if sufficient flux is used, there will be voids in the body. Voids may weaken the binding of the crystalline phase in the body and will facilitate the vibration of the small particles under the influence of an applied electric field, thus generating a sound wave. These voids can be minimized by proper technique in the sample preparation.

In the aforementioned cases, the dielectric losses are functions of the elastic properties of the ceramic as well as the dielectric properties of the component phases. Experimental tests for examining the relation between the dielectric loss and the elastic property of ceramic bodies are necessary. The result is not available at present but will be undertaken in the near future.

Wollastonite was found to be an excellent crystalline phase for ultra low loss bodies in the earlier work.[4] The dielectric losses of such wollastonite-type bodies are next considered as a function of the wollastonite content.

Figure 2(*A*) is an assumed diagram of the dielectric loss of a series of wollastonite bodies. The upper dot-dash line represents the loss due to the wollastonite crystals and the matrix phase (glass and lead aluminum silicate). The approximate shape of the curve may be estimated by using Nicsel's equation.[3] When the ratio of the dielectric constants of the wollastonite crystals and the matrix phase is nearly 1, the curve can be approximated by a straight line. The broken line represents the dielectric loss caused by the voids included in the body. This loss would increase rapidly with decreasing flux content because the lack of flux will introduce many voids. The upper solid line represents the resultant loss which might have a minimum value at a certain wollastonite concentration.

Assuming that the body has the proper amount of flux and has been properly fabricated, the dielectric loss due to voids should be very small. Therefore, the main part of the dielectric loss of the ceramic body will be due to the wollastonite crystals and the matrix phase. Since the dielectric loss of the wollastonite crystal is fixed, probably the best way to decrease the dielectric loss of the body is to use a matrix with a lower dielectric loss. In this case, the resultant dielectric loss will also be lowered (as shown by the lower set of curves in Fig. 2(*A*)). But, if the dielectric loss of the matrix phase were lower than that of the wollastonite crystals, then the dielectric loss vs. wollastonite concentration curve would not have a minimum, as in Fig. 2(*B*). In such case, it is better to use glass itself as the dielectric, and there would be no need to use wollastonite if the purpose is to make the lowest dielectric loss body.

Since the matrix phase of the body is a glass or a mixture of a glass and a small amount of little crystals, the theoretical dielectric loss of glasses in relation to composition can be correlated with the resultant dielectric loss of ceramic bodies.

Most glasses contain silicon-oxygen networks as the main structural units and several kinds of positive ions. Usually, some of the positive ions are bound to Si-O networks with rather weak forces which may be of ionic or covalent character. Because the distribution of Si-O networks in the glass lacks an orientational regularity, there must be many potential minima for the positive ions. It can be assumed that

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[1] Karl Lichtenecker, "Dielectric Constant of Natural and Synthetic Mixtures," *Physik. Z.*, **27**, 115-58 (1926).

[2] D. A. G. Bruggeman, "The Calculation of Various Physical Constants of Heterogeneous Substances: I, Dielectric Constants and Conductivities of Mixtures Composed of Isotropic Substances," *Ann. Phys.*, **24**, 636-64 (1935).

[3] W. Nicsel, "Energy Distribution in Mixtures of Two Dielectrics in an Electric Field and Calculations of the Loss Angle in Quasistationary Processes," *Ann. Phys.*, **12**, 410 (1953).

Table I. Polarizability of Several Ions According to Pauling

	(× 10^{-24} cm.^{3})					
Li^+	0.029	Be^{2+}	0.008	B^{3+}	0.003	C^{4+}	0.0013
Na^+	0.179	Mg^{2+}	0.094	Al^{3+}	0.052	Si^{4+}	0.0165
K^+	0.83	Ca^{2+}	0.47	Sc^{3+}	0.295	Ti^{4+}	0.185
Rb^+	1.40	Sr^{2+}	0.86	Y^{3+}	0.55	Zr^{4+}	0.87
Cs^+	2.42	Ba^{2+}	1.55	La^{3+}	1.04	Ce^{4+}	0.73
O^{--}	3.88						

Table II. Polarizabilities of Ions According to Roberts*

	(Cubic angstroms)					
F^-	25.8	Li^+	11.7	Mg^{2+}	11.4	Si^{4+}	2.2
Cl^-	62.8	Na^+	30.2	Ca^{2+}	34.0	Ti^{4+}	34.5
Br^-	81.0	K^+	42.8	Sr^{2+}	49.1		
I^-	108.4	Rb^+	56.1	Ba^{2+}	70.6		
O^{--}	30.9	Cs^+	71.3	Pb^{2+}	91.8		
		Cu^+	27.2				
		Ag^+	29.4				
		Tl^+	80.1				

* This table shows the high polarizabilities of Pb^{2+}, Ba^{2+}, Tl^+, and Cs^+.

the positive ions are distributed in many potential wells according to the laws of statistical mechanics.

The potential energy of a positive ion with a charge e is given by the equation

$$W = e\Phi - \tfrac{1}{2}\alpha E^2$$

Φ (must be negative) = electric potential.
E = electric field strength, at the position of the ion.
α = polarizability of the ion.

Imagine a case when Na ions of Na glass are replaced by Pb or Ba ions. Since the electric charge of Pb (or Ba) ion is twice that of Na ion and the polarizability is about 3 or 4 times that of Na ion, the depth of the potential well for Pb (or Ba) ion would be more than twice that of Na ion, assuming that Φ and E are of the same value for the two glasses. Therefore, the deep potential wells can be realized if the valency and polarizability of the ions are large. It will be noted in Table I that Ba and Cs have high electronic polarizabilities.

Roberts[1] has calculated polarizabilities of ions in simple crystals. The basic equation from which the polarizabilities have been calculated is

$$\alpha = \Sigma\alpha_i = 3V(K - 1)/(K + 2)$$

V = volume of the unit cell in the crystal lattice divided by the number of molecules which it contains.
α = polarizability per molecule.
α_i = polarizability of the respective ions.
K = dielectric constant of the crystal.

Using the observed data of K of a number of crystals, the polarizabilities have been estimated and are tabulated in Table II.

The foregoing equation is based on the m.k.s. system; to obtain the polarizabilities based on the c.g.s. unit system, the polarizabilities tabulated above should be divided by 4π. It should be noted that the polarizabilities obtained by Roberts[1] include not only the electronic parts but also the ionic parts. The electronic polarizabilities can be assumed to have approximately a constant value irrespective of the crystal structure in which the ion exists, but the ionic polarizabilities should change depending on the crystal structure.

[1] Shephard Roberts, "Dielectric Constants and Polarizabilities of Ions in Simple Crystals and Barium Titanate," *Phys. Rev.*, 76, 1215–20 (1949).

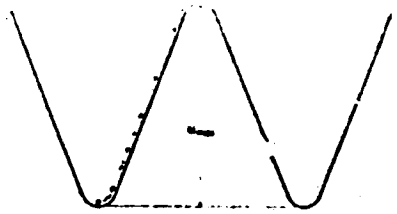

Fig. 3. Probability of escape of positive ions from one interstice to another.

The structure of glass is more complex than most crystals so that the polarizabilities of ions in the glass should be different from those listed in Tables I and II. Nevertheless, it can be assumed that the orders of magnitude are the same even though the ions are in the crystalline or in the glassy state.

When an electric field is applied to the glass, dielectric polarization, which is composed of electronic and ionic polarization, will appear.

$$P = P_{elec} + P_{ion}$$

$$P_{ion} = P'_{ion} + P''_{ion}$$

where P_{elec} is due to the distortion of the electron cloud of the ion by the applied field; P_{ion} is due to the shifting of the ions; one part of which (P'_{ion}) is due to the small shift in each potential well and another part of which (P''_{ion}) is due to the rearrangement of the distribution of all the positive ions over all the potential wells under the influence of the applied electric field. Since the probability of escape of one positive ion from the potential well is proportional to the exp $[-(U'_{max}/kT)]$ (Fig. 3), P''_{ion} must depend strongly on the depth of this potential well and the temperature. If the binding force between the positive ion and the Si-O network is sufficiently strong so that the potential well for the positive ion is very deep, then the dielectric loss due to P'_{ion} appears only at very high frequency, such as infrared-optical frequency, because the restoring force for the shift of the ion is strong in the deep potential well. Dielectric loss due to P''_{ion} may appear at very low frequency but disappears at high frequency (intermediate radio frequency range). The reason is as follows: Since the rate of transfer of the positive ion from one potential well to another is proportional to the exp $[-(U'_{max}/kT)]$, then the rate is smaller for deeper potential wells and lower temperature. If the rate of transfer is small and the frequency of the applied field is high, the positive ions do not have sufficient time to reach the state of equilibrium distribution which is rapidly changing with the a.c. electric field. As a result, there is almost no contribution from P''_{ion} when the frequency is sufficiently high. In such cases, P'_{ion} and P''_{ion} contribute little to the dielectric loss in the radio frequency range.

The following example shows the way the depth of the potential well affects the dielectric loss at different frequencies. It has been stated that the loss due to P''_{ion} appears at approximately the frequency given by the equation

$$\omega_0 = \omega_1 \exp\left(-\frac{2U'_{max}}{kT}\right)$$

where ω_1 is $\sim 10^{13}$ sec.$^{-1}$. Assume that a certain Na glass shows a high dielectric loss at $\omega_0 = 10^8$ sec.$^{-1}$ (about 15 mc.). If the Na ion is replaced by Pb or Ba ion and the potential depth becomes twice that of the Na glass, then for the Pb or

Table III. Behavior of ϵ_w and Tan δ at Different Frequencies

	ω^*	ωT_1	ωT_1^2	ϵ_w	$\frac{\omega \epsilon_0 T_1}{1+\omega^2 T_1^2}$
High frequency	$\frac{1}{T_1}$	1	1	$\epsilon_\infty + \frac{\epsilon_0}{2}$	$\frac{\epsilon_0}{2}$
	$10\frac{1}{T_1}$	10	100	$\epsilon_\infty + \frac{\epsilon_0}{101}$	$\frac{10\epsilon_0}{101} \sim \frac{\epsilon_0}{10}$
	$100\frac{1}{T_1}$	100	10,000	$\epsilon_\infty + \frac{\epsilon_0}{10001}$	$\frac{100\epsilon_0}{10001} \sim \frac{\epsilon_0}{100}$
Low frequency	$\frac{1}{10}\frac{1}{T_1}$	0.1	0.01	$\epsilon_\infty + \frac{\epsilon_0}{1.01}$	$\frac{0.1\epsilon_0}{1.01} \sim \frac{\epsilon_0}{10}$
	$\frac{1}{100}\frac{1}{T_1}$	0.01	0.001	$\epsilon_\infty + \frac{\epsilon_0}{1.0001}$	$\frac{0.01\epsilon_0}{1.0001} \sim \frac{\epsilon_0}{100}$

* ω will contribute to ϵ_w when $\omega \leq (1/T_1)$ $\epsilon_w = \epsilon_\infty + \epsilon_0$, not when $\omega > (1/T_1)$ $\epsilon_w = \epsilon_\infty$.
ω will contribute to tan δ when ω is approximately $1/T_1$ but will not contribute further from $1/T$ than one decade.

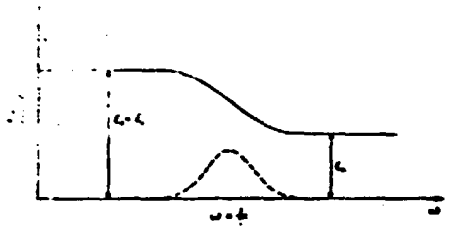

Fig. 4. Behavior of ϵ_w and tan δ at different frequencies.

Table IV. Power Factors of Several Glasses According to J. M. Stovels

	Power factor*
Pure fused SiO_2 (SiO_2 100%)	0.0003
Na glass (SiO_2 70.4%; Al_2O_3 2.3%; Na_2O 12.0%; CaO 8.8%)	0.0096
Ba glass (SiO_2 30.0%; B_2O_3 10.0%; Al_2O_3 10%; BaO 50.0%)	0.0013
Pb glass (SiO_2 37.5%; Al_2O_3 2.0%; Na_2O 8.5%; K_2O 2.0%; PbO 54.0%)	0.0010

* 1.5 mc., 30°C.

Ba glass, ω_0 is given by the following equation, assuming ϵ_0 is the same,

$$\omega_0 = \omega_1 \exp\left(-\frac{U_{max}}{kT}\right)$$

resulting in $\omega_0 = 10^{-11}$ which is a very low frequency. Consequently, there must be almost no dielectric loss due to P_{max} over the frequency range from about 100 c.p.s. to about 10^{11} c.p.s. Therefore, glasses containing deep potential wells can be expected to show a maximum dielectric $\epsilon \tan \delta$, on the loss vs. frequency curve, at a low frequency. At about the frequency range where the dielectric constant begins to decrease (Fig. 4), the phase of P'_{max} is delayed compared to the phase of the applied field. As a result, high dielectric loss appears. Generally speaking, when the dielectric constant, ϵ, for low frequency is appreciably higher than the square of the refractive index n for optical frequency, a dielectric loss should be expected due to P_{max} at a certain frequency range where $d\epsilon/d\omega$ is large; ω is the frequency of the a.-c. field.

The following illustration (Table III, Fig. 4) shows the behavior of the dielectric constant and power factor at different frequencies.

$$\epsilon_w = \epsilon_\infty + \frac{\epsilon_0}{1+\omega^2 T_1^2}$$

$$\tan \delta = \frac{1}{\epsilon_w} \cdot \frac{\omega \epsilon_0 T_1}{1+\omega^2 T_1^2}$$

ϵ_w = dielectric constant at any applied frequency
ϵ_∞ = dielectric constant immediately following the applied field
ϵ_0 = dielectric constant after the application of a constant field.
T = relaxation time.

The foregoing equation is for only one relaxation time. When there is more than one relaxation time

$$\epsilon_w = \epsilon_\infty + \frac{\epsilon_0}{1+\omega^2 T_1^2} + \frac{\epsilon_0'}{1+\omega^2 T_2^2} + \frac{\epsilon_0''}{1+\omega^2 T_3^2} \text{ etc.}$$

Actual glasses contain many kinds of potential wells with various potential depths. The explanation given above, however, can explain qualitatively the low loss character of glasses containing deep potential wells.

Examples of dielectric loss for several glasses are shown in Table IV. It is clear from these data that glasses containing ions such as Pb or Ba, which have high electronic polarizabilities, show appreciably lower dielectric loss than does Na glass. Pure fused silica does not contain foreign ions so that dielectric loss due to ionic movement should be very low. When a glass contains foreign ions, higher valency, as well as higher electronic polarizability of the ions, is necessary for the glass to have a low dielectric loss.

(f) *Summary of Theoretical Analyses*

When a ceramic consists of two or more phases, the principal dielectric loss is due to the dielectric loss of the component phases and may be due also to the elastic loss of the ceramic body. To decrease the loss, the following conditions are necessary:

(1) The dielectric losses of the component phases (crystal phase and binder phase) should be as low as possible. A low dielectric loss of the binder phase can be realized if it contains ions such as Pb, Bi, Tl, Cd, and Ba, which have high valency and high electronic polarizability.

Table V. Body Compositions (wt. %) and Properties

	ES-1	ES-2	ES-3	ES-4	ES-5	ES-6
No. 4248 wollastonite	60.0	55.0	60.0	60.0	55.0	60.0
No. 4 ball clay	20.0	20.0	10.0	20.0	20.0	10.0
Barium carbonate	10.0	15.0	15.0	10.0	15.0	15.0
Lead bisilicate	10.0	10.0	15.0			10.0
Bismuth trioxide				5.0	5.0	5.0
Silica				5.0	5.0	
Maturing temp. (°F.)	2100	2100	2100	2100	2100	2100
Water absorption	0.00	0.00	0.00	0.00	0.00	0.00
Power factor (%) (at 1 mc. and room temp.)*	0.018	0.008	0.017	0.014	0.013	0.014
Dielectric constant*	6.85	6.88	7.62	6.92	7.15	7.40
Loss factor (%)*	0.122	0.056	0.130	0.101	0.092	0.108
JAN-1-10 grade	L-7	L-8	L-7	L-7	L-8	L-7

* These values are average results of at least 10 specimens.

(2) Reduce the loss due to elastic causes. For this purpose, it is suggested that (a) a nonpiezoelectric material be used as the principal component of the body; (b) the principal dielectric constants ϵ_1, ϵ_2, and ϵ_3 of the principal crystalline phase should be nearly the same and they should be almost equal to the dielectric constant of the binder phase; (c) if the difference of the dielectric constants of the phases is relatively large, the grain of the principal phase should be nearly spherical in shape; (d) the glassy or binder phase should cement the crystals tight enough to prevent a conversion of electric energy to mechanical energy; and (e) the body must be as dense as possible.

(3) Cool the ceramic body very slowly, the reason being the same as for the annealing of a glass. During the annealing process, rearrangement of molecular networks or ions in the glass takes place resulting in more stable configurations of the networks and the ions. In this case, the potential wells for the ions can be expected to be deeper than those of the unannealed glass.

III. Experimental

(1) Preparation of Specimens

A 1000-gm batch of the composition was weighed, and 2000 cc. of water were added; the batch was wet ball milled for 24 hours and then dried completely. The dried batch was passed through a micropulverizer, mixed in a Lancaster mixer for 10 minutes while adding 6% water by weight, and then mixed for an additional 20 minutes in the Lancaster. The material thus prepared was pressed into disks 2 in. in diameter to provide close contact of particles and to initiate preliminary chemical reactions during a prefiring to 1600°F. The pressed disks were prefired to eliminate the voids caused by gaseous evolution from the components and to minimize internal stresses, which would otherwise develop in the grain boundaries, in the final test specimens.

The prefired disks were crushed and dry ball milled for about 15 hours to pass a 200-mesh screen. Six cc. of Superhoid binder (1.5% solution) were added per 100 gm. of powder and mixed in a Lancaster mixer for 30 minutes. The powder was pressed into specimens (disks approximately 2 in. in diameter and 0.1 in. thick) at 7000 lb. per sq. in., fired to the maturing temperature in 4 hours, soaked at that temperature for 30 minutes, and cooled very slowly to room temperature during approximately 19 hours.

(2) Experimental Bodies

Several experimental bodies were designed based on the previous theoretical discussion. The compositions of some of these bodies are given in Tables V and VI.

(3) Test Methods

Dielectric constants and power factors were measured at 1 mc. and room temperature using a Boonton *Q* meter.

Silver paint was used as the electrodes to provide a better contact with the material because it can flow into the minute depressions on the surface of the specimen. Care was taken to paint on enough silver so that there was no resistance on the surface, measured between the opposite edges of the specimen. The silvered specimens were dried slowly on a hot plate for 30 minutes to about 500°F. and held there for 1 hour to make sure that all the organic solvents of the silver suspension were evaporated. The specimens were cooled to room temperature in a desiccator.

During the entire electrical test period, the humidity and temperature must be fairly constant to obtain consistent results. The conditions (temperature and humidity) can be assumed to be constant when the capacitance and *Q* readings with air as the dielectric remain the same.

All of the matured bodies, including those shown in Tables V and VI, were analyzed by X-ray and petrographic methods.

IV. Discussion of Results

Referring to Table V, bodies ES-1, ES-2, and ES-3 were fluxed with lead and barium, bodies ES-4 and ES-5 were

Table VI. Body Compositions (wt. %) and Properties

	JK-1	JK-2
No. 4248 wollastonite	60.0	60.0
Frit JS-1	40.0	
Frit JS-2		40.0
Maturing temp. (°F.)	2000	2000
Water absorption	0.00	0.00
Power factor (%) (at 1 mc. and room temp.)*	0.0734	0.0273
Dielectric constant*	6.94	7.38
Loss factor (%)*	0.502	0.193
JAN-1-10 grade	L-6	L-7

Frit Compositions

	JS-1	JS-2
PbO	54.0	54.0
SiO_2	37.5	37.5
Al_2O_3	2.0	2.0
Na_2O	3.5	
K_2O	3.0	
BaO		6.5

*These values are average results of at least 10 specimens.

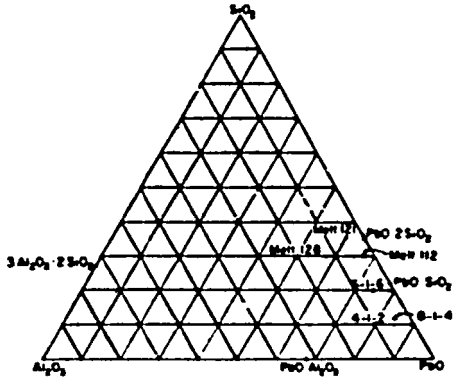

Fig. 5. Ternary compounds in the high lead oxide portion of the system $PbO-Al_2O_3-SiO_2$.

fluxed with bismuth and barium, and body ES-6 was fluxed with lead, bismuth and barium. All of these bodies were found to have ultra low dielectric loss (L-7 and L-8, JAN-1-10); they possessed heavy modifying cations such as Pb^{++}, Ba^{++}, and Bi^{+++}, which have high valency (in comparison with the alkalis) and high electronic polarizabilities. This conforms to the previous theoretical explanation of deep potential wells.

Table VI gives two body compositions in which all of the glass phases were prefritted. The compositions of the frits are also given in Table VI. These two bodies provide a clear comparison of the effect of heavy cations with high electronic polarizability on dielectric loss.

The glass phase of JK-1 contains Na_2O and K_2O and the dielectric loss is relatively high ($\tan \delta = 7.2 \times 10^{-4}$). The presence of alkali ions increases the power loss. Where BaO replaces the alkalis, as in JK-2, the dielectric constant is somewhat increased. The power factor, however, is substantially reduced ($\tan \delta = 2.7 \times 10^{-4}$ for JK-2). It appears that lead and barium together provide the best fluxing system of those tried to date for ultra low loss ceramic bodies.

The X-ray patterns show that all of the bodies contain β-wollastonite crystals. Bodies ES-1, ES-2 ES-3, and ES-6 show a few other peaks identified as lead aluminum silicate, which is melt 128 in the ternary system $PbO-Al_2O_3-SiO_2$ (Fig. 5). The calculated batch composition of melt 128 was fabricated into a ceramic body. The body has a firing range of 1300° to 1460°F. and shows a self-glazing property. Bodies ES-4 and ES-5, which were fluxed with bismuth instead of lead, show the same peaks as the other ES bodies. No report was found in the literature of any bismuth compounds that coincide with these peaks. A composition similar to melt 128, but using Bi_2O_3 instead of PbO, was fabricated and fired to maturity at 1600°F. The fired body was analyzed by X-ray and showed the same peaks as melt 128. Probably bismuth reacts the same way as lead and forms bismuth aluminum silicate similar in structure to lead aluminum silicate.

Thin sections of the bodies were examined petrographically and were found to contain crystals, glass, and small amounts of voids. The crystals, which were small and of irregular shape, were identified as lead aluminum silicate by X ray. The wollastonite crystals cannot be seen without crossed-Nicols because the refractive index of wollastonite is very close to that of the immersion medium.

Using crossed Nicols some of the predominantly larger wollastonite crystals are very clearly seen. The remaining tiny crystals are a mixture of wollastonite crystals and lead aluminum silicate crystals.

It can be noted that although the fired bodies contain wollastonite crystals which have needlelike shapes, they have ultra low dielectric loss. These may be explained by the following: (1) The dielectric constants of the component phases are relatively close (ϵ of wollastonite crystal = 6.15; ϵ of bodies = 7.14; consequently the ϵ of the binder phase is approximately 8.2). (2) The wollastonite crystals are well bonded by the tiny crystallites, which have self-glazing property, and the glass phase. Therefore, the elastic loss due to vibration of the wollastonite crystals may be very small.

V. Conclusions

(1) Experimental bodies, which were designed on the basis of theoretical considerations, provide for ultra low dielectric loss ceramics.

(2) The ultra low loss ceramic body should be composed of a low loss principal crystalline phase such as wollastonite with a binder phase containing heavy modifying cations with high electronic polarizabilities such as Pb^{++}, Ba^{++}, and Bi^{+++}. It appears that Pb and Ba together provide the best fluxing system of those tried to date for ultra low dielectric loss ceramics.

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