

Dielectric Constant and Loss Angle of Several Solid Dielectrics at a Wavelength of 3 cm, and Their Temperature and Frequency Dependence

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The results of measurements of ϵ and $\tan\delta$ at a wavelength of 3 cm are given for magnesium, zinc, calcium, strontium and bismuth titanates, barium tetratitanate and steatite. A cylindrical resonator was used. The frequency dependences of ϵ and $\tan\delta$ were determined for these materials over a wide range of frequencies, and the temperatures were determined at frequencies of 10^3 , 10^6 and 10^{10} cps. The results are discussed from the point of view of present ideas concerning the dielectric constant.

1. STATEMENT OF THE PROBLEM

It is known^{1,2} that the dielectric constant of polycrystalline dielectrics of the ceramic type may have different values, lying in a wide range (from 6 to several thousand), depending on their composition and structure. High values of ϵ for nonpiezoelectric titanates of the perovskite and rutile structure depend on a favorable internal field³. On introducing weakly bound ions into such dielectrics, in a manner dependent on the structural defects, for example, a thermal ionic relaxation polarization is produced which strongly increases the dielectric constant (up to 1000)³ and leads to a rather strong frequency and temperature dependence of ϵ and $\tan\delta$.

The indicated dependence agrees satisfactorily with the results of relaxation theory, provided the internal field is taken into account correctly⁴. In the further development of the theory of polarization of solid dielectrics, the investigation of their dielectric properties in ultrahigh frequency region and in the long wavelength part of the infrared region of the spectrum will be of very great significance. Actually, in dielectrics with purely elastic polarization the dielectric constant should not depend on the frequency until the frequency of the ions in the crystal lattice ($10^{12} - 10^{13}$ sec⁻¹). In dielectrics with relaxation polarization a decrease in ϵ with frequency should take place at lower frequencies and can arise at room temperature in the centimeter wavelength region. Suggestions as to the existence of a suitable dispersion for piezoelectrics, for example, are to be found in the literature⁵.

In the infrared region of the spectrum all crystals, as is well known, display anomalous dispersion and absorption. The lower the frequency corresponding to maximum absorption, the greater must be the elastic ionic polarizability. Generally

speaking, a high value of the dielectric constant can depend not only on a suitable internal field and, in the case of dielectrics with relaxation polarization, increased thermal ionic (or dipole) polarizability, but also on a high value of elastic polarizability.

Dielectric losses in solid dielectrics have been investigated by many authors. In the most recent works^{6,7}, it has been shown that more or less clearly manifested dielectric losses are superimposed on the loss of conductivity even in simple ionic crystals. In these dielectrics, in which relaxation polarization leads to a high value of the dielectric constant, dielectric losses are ordinarily comparatively large, and the loss angle has temperature and frequency maxima in the region of electrotechnical and radio frequencies. Dielectrics with elastic polarization ordinarily have small losses. In a rather wide frequency interval the loss angle of these dielectrics does not depend on the frequency and begins to increase with temperature only in the region of comparatively high temperatures. The mechanism of these small dielectric losses is still insufficiently clear. On going to ultrahigh frequencies, the loss angle of solid dielectrics with elastic polarization may increase if the region of resonance absorption corresponds to the long wavelength part of the infrared region of the spectrum.

The investigation of dielectrics in the centimeter wavelength region also has a practical value in connection with their use in high frequency techniques.

As a rule, the works mentioned involve great procedural difficulties, the surmounting of which is possible only by use of good ultrahigh frequency and special optical apparatus, and development and utilization of new methods are needed.

In the present work, ultrahigh procedures published in the literature⁵ are used with certain small

but essential improvements, allowing the measurement of the dielectric constant of solid dielectrics over a rather wide range of values (from 1.5 to 200) at a wavelength of 3.1 cm.

For the measurements at audio and radio frequencies an audio bridge of type 716-V and Q meters of types KV-1 and UK-1 were used.

2. METHODS OF MEASUREMENT AT 3.1 CM WAVELENGTH

The basic part of the ultrahigh frequency apparatus is the circular cylindrical resonator, operating in the H_{01} mode. This mode is excited through two openings in the end wall of the cavity joined to a thin wall of a rectangular waveguide in which the wave is propagated⁸. By a careful choice of the dimensions, form and position of the output loop in the coupler, we were able, first, to obtain a signal in the basic mode H_{01} exceeding by 200-400 times the signals in other modes which were unavoidably present in the resonator, and, second, to obtain a loaded resonator Q approaching that of the resonator itself, that is, to make the energy losses in the coupler negligibly small in comparison with the energy losses in the walls of the cavity. The resonator was constructed of brass and was silvered. Its diameter was 45 mm. The Q of the empty resonator for the H_{01} mode is 20,000. The samples being measured are prepared in the form of disks of diameter 1 mm less than that of the cylinder, but the optical thickness depends on the magnitudes of ϵ and $\tan\delta$. The thickness error in the preparation of the disks is not more than 0.01 mm. The disks are set tightly against the electrically unconnected movable plunger serving as the second end wall of the cavity. For good contact of the samples with the plunger, the resonator is positioned vertically. The samples are placed in the resonator through a cut in the side wall located below the working positions of the plunger. The plunger is moved with the aid of a micrometric screw with steps of 1 mm and a distance of travel of 100 mm. The position of the plunger is read on a scale, shown in Fig. 1, with an accuracy of 0.01 mm; this accuracy is assured by the quality of the micrometric screw.

The changes in the resonance length and Q of the cavity which occur when a dielectric is placed in it allow the calculation of ϵ and $\tan\delta$ for the sample under investigation. The scheme of the calculation is given in detail in reference 5 and briefly in reference 8. In the calculation it is assumed that the propagation constant of the electromagnetic wave in the dielectric under investigation $\gamma = \alpha + j\beta \approx j\beta$, that is, that $\alpha \ll \beta$. A block

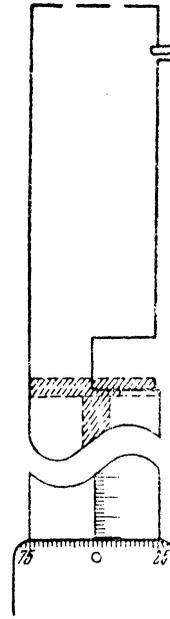


FIG. 1. The cavity resonator.

diagram of the apparatus is shown in Fig. 2; it contains five assemblies: 1, a generator of type 43-I; 2, the measuring resonator; 3, a wavemeter; 4, an amplifier-mixer; 5, a cathode ray oscillograph of type EO-7. In addition, in order to isolate the resonator from the wavemeter, there is introduced into the rectangular waveguide 6 an attenuator 7. The modulation is fed into the reflex klystron oscillator through a condenser and potentiometer from the horizontal sweep of the oscillograph.

The precision wavemeter is in the form of a cylindrical cavity resonator, excited, like the measuring resonator, through two openings in an end wall and operating in the H_{01} mode. The signal for the measurements is taken through an output coupler in the side wall, exciting the H_{01} mode in a rectangular waveguide. The signal taken out through the coupler into the rectangular waveguide is detected and fed to the input of the amplifier-mixer. The peak value of the intensity of the signals from the resonator and wave meter applied to the input of the amplifier is about 0.2 mv. The amplifier-mixer contains three amplifiers in cascade: two single 6Zh6 amplifiers, into one of which is fed the signal from the resonator and into the other of which is fed the signal from the wavemeter, and a single 6N9 mixer. The amplification factor is about 2000. The pass band includes all of 50-20,000 cps and assures the preservation of the form of the signals from the resonator and the wavemeter. The horizontal sweep of the oscillograph (with a frequency of 50 cps) appears as a frequency

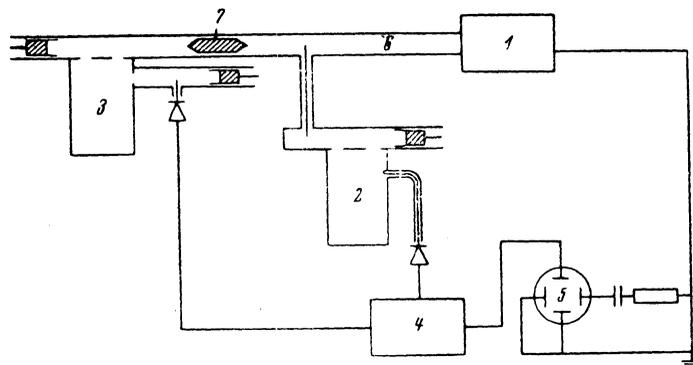


FIG. 2. Block diagram of the apparatus: 1, generator; 2, resonator; 3, wavemeter; 4, amplifier; 5, oscillograph; 6, waveguide; 7, attenuator.

in the resultant display; hence with a quadratic detector the width of the resonance curve obtained on the oscillograph screen gives the width of the resonance curve of the resonator, which may be measured with the aid of the wavemeter. The wavemeter allows increasing the accuracy of the determination of the resonance-length setting of the resonator when a sample is put into it even with poor generator stability, since the wavemeter fixes the working frequency with great accuracy.

In taking the temperature characteristics of the samples, experimentally determined corrections are applied to the Q of the resonator and to the other measurements which are changed by heating. The resonator is heated by means of a heating spiral surrounding its side walls. The temperature of the sample is determined by a thermocouple placed in a hole in the plunger next to the lower surface of the sample. An estimate of the relative error in the determination of ϵ and $\tan \delta$ at room temperature gives ± 1 to 1.5% and $\pm 15\%$, respectively, for all the values of ϵ and $\tan \delta$ mentioned below. The average relative errors in ϵ and $\tan \delta$ found from the experimental spread in the values of ϵ and $\tan \delta$ for various samples did not, as a rule, exceed these values. At elevated temperatures the error in the determination of $\tan \delta$ was increased threefold.

3. RESULTS OF THE MEASUREMENTS

The values of the dielectric constant and loss angle at a wavelength of 3.1 cm are given for a number of substances in Tables 1-8. From these tables it is clear that for a correct choice of sample thickness, results are obtained which are reproducible and lie close together for different samples.

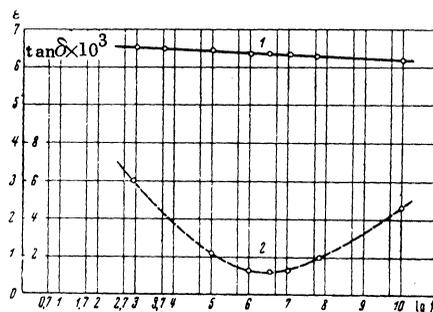


FIG. 3. Frequency dependence of ϵ and $\tan \delta$ for steatite. Curve 1 gives ϵ and curve 2 gives $\tan \delta$.

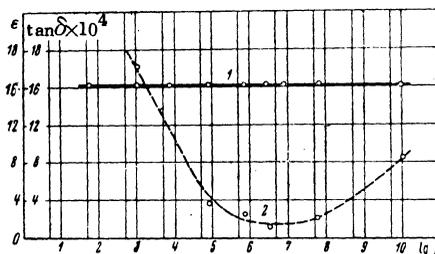


FIG. 4. Frequency dependence of ϵ and $\tan \delta$ for magnesium titanate to which a small amount of calcium titanate and flux has been added. Curve 1 gives ϵ and curve 2 gives $\tan \delta$.

Curves showing the dependence of ϵ and $\tan \delta$ on frequency over a wide range of frequencies are given in Figs. 3-9. Practically complete absence of any frequency dependence of the dielectric constant is found for magnesium titanate, barium titanate, zinc titanate and fused strontium titanate (Figs. 4, 5, 6, 9). This fact fully confirms our

idea that only elastic polarization occurs in these materials (the question as to whether strontium titanate belongs to the piezoelectrics seems open to discussion).

In the case of the frequency dependence of stearite and calcium titanate, it is clear from Figs. 3 and 8 that with increasing frequency a rather weak, almost linear decrease in the dielectric constant takes place. This could not occur for purely elastic polarization. It is possible that in these materials weakly expressed relaxation processes occur. The dielectric constant of bismuth titanate decreases rather strongly with frequency in the region of low frequencies (Fig. 7). This can be explained only by the presence of relaxation processes with long relaxation times.

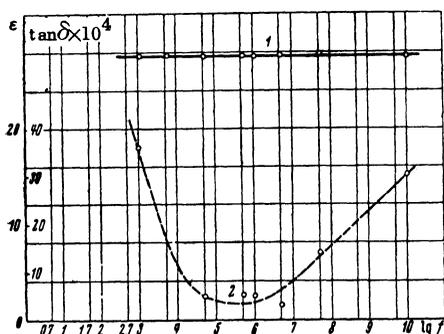


FIG. 5. Frequency dependence of ϵ and $\tan \delta$ for fused barium titanate. Curve 1 gives ϵ and curve 2 gives $\tan \delta$.

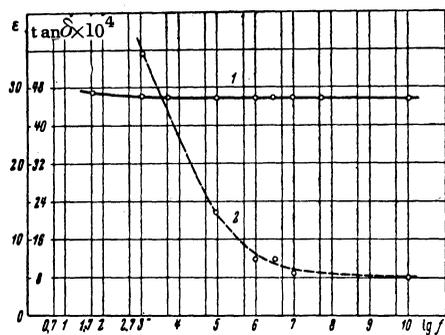


FIG. 6. Frequency dependence of ϵ and $\tan \delta$ for zinc titanate. Curve 1 gives ϵ and curve 2 gives $\tan \delta$.

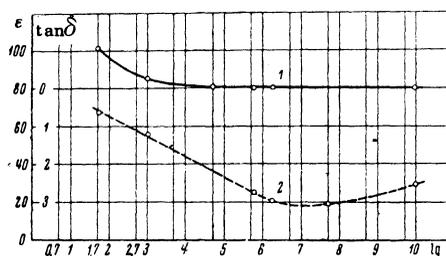


FIG. 7. Frequency dependence of ϵ and $\tan \delta$ for bismuth titanate. Curve 1 gives ϵ and curve 2 gives $\tan \delta$.

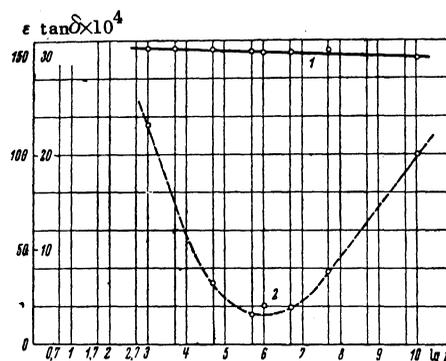


FIG. 8. Frequency dependence of ϵ and $\tan \delta$ for calcium titanate. Curve 1 gives ϵ and curve 2 gives $\tan \delta$.

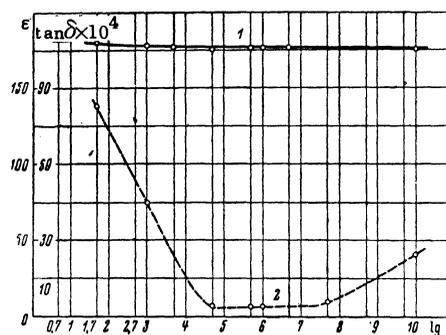


FIG. 9. Frequency dependence of ϵ and $\tan \delta$ for fused strontium titanate. Curve 1 gives ϵ and curve 2 gives $\tan \delta$.

The dielectric loss angle of all the dielectrics investigated, with the exception of zinc titanate, shows an unsymmetrical but definite minimum in the frequency region 10^5 - 10^8 cps. The increase in the loss angle at low frequencies is ordinarily connected with conductivity losses, for which the loss

TABLE 1

Material	Sample no.	Sample thickness in mm	ϵ	$\tan \delta$	Average ϵ	Average $\tan \delta$
Steatite	1	2.19	6.51	0.0023	6.50	0.0026
	2	2.11	6.51	0.0027		
	3	2.41	6.49	0.0025		
	4	2.40	6.51	0.0026		
	5	2.30	6.46	0.0028		
	6	2.50	6.51	0.0024		
	7	2.60	6.48	0.0032		

TABLE 2

Material	Sample no.	Sample thickness in mm	ϵ	$\tan \delta$	Average ϵ	Average $\tan \delta$
Magnesium titanate	1	1.79	16.1	0.00077	16.0	0.00086
	2	1.79	16.2	0.00094		
	3	1.78	16.0	0.00099		
	4	2.37	15.9	0.00082		
	5	1.80	15.8	0.00086		
	6	2.23	16.1	0.00079		
	7	2.23	16.0	0.00069		
	8	2.23	15.9	0.00077		

TABLE 3

Material	Sample no.	Sample thickness in mm	ϵ	$\tan \delta$	Average ϵ	Average $\tan \delta$
Barium tetra-titanate	1	1.88	28.1	0.0031	27.6	0.0030
	2	2.00	27.9	0.0028		
	3	2.00	27.7	0.0030		
	4	3.76	27.8	0.0028		
	5	1.73	27.7	0.0027		
	6	1.75	27.4	0.0031		
	7	1.29	27.5	0.0033		
	8	1.29	27.4	0.0032		
	9	1.29	28.0	0.0033		
	10	1.75	27.3	0.0030		
	11	1.75	27.9	0.0028		
	12	1.75	27.2	0.0030		

TABLE 4

Material	Sample no.	Sample thickness in mm	ϵ	$\tan \delta$	Average ϵ	Average $\tan \delta$
Zinc titanate	1	4.70	27.8	0.00090	27.9	0.00079
	2	4.70	27.9	0.00064		
	3	4.70	27.9	0.00078		
	4	1.70	28.0	0.00074		
	5	1.80	27.7	0.00090		

TABLE 5

Material	Sample no.	Sample thickness in mm	ϵ	$\tan \delta$	Average ϵ	Average $\tan \delta$
Calcium titanate	1	3.30	152	0.0020	152	0.0020
	2	3.30	152	0.0020		
	3	1.80	151	0.0022		
	4	4.30	152	0.0015		
	5	4.30	151	0.0022		
	6	1.81	151	0.0019		

TABLE 6

Material	Sample No.	Sample thickness in mm	ϵ	$\tan \delta$	Average ϵ	Average $\tan \delta$
Bismuth titanate	1	2.82	81.0	0.0031	80.8	0.0033
	2	2.82	81.0	0.0028		
	3	2.82	80.0	0.0026		
	4	3.95	81.5	0.0034		
	5	2.82	83.0	0.0043		
	6	3.81	80.3	0.0037		
	7	3.80	80.7	0.0033		
	8	3.80	79.8	0.0032		

TABLE 7

Material	Sample No.	Sample thickness in mm	ϵ	$\tan \delta$	Average ϵ	Average $\tan \delta$
Strontium titanate	1	3.90	175	0.0023	176	0.0025
	2	4.99	177	0.0021		
	3	1.60	176	0.0031		
	4	4.99	176	0.0025		
	5	4.99	172	0.0031		
	6	2.75	178	0.0024		
	7	4.26	175	0.0019		
	8	4.26	175	0.0026		
	9	1.95	175	—		

TABLE 8

Material	Sample No.	Sample thickness in mm	ϵ	$\tan \delta$	Average ϵ	average $\tan \delta$
Porcelain	1	2.50	5.47	0.0084	5.39	0.0113
	2	2.60	5.34	0.0101		
	3	2.41	5.35	0.0104		
	4	2.39	5.42	0.0142		
	5	2.40	5.39	0.0135		

angle is inversely proportional to frequency. However, it is possible to consider that in the cases where the dielectric constant decreases with frequency in the region of large values of $\tan \delta$, relaxation polarization occurs. This polarization, which is especially strongly manifested in bismuth titanate takes place, perhaps, in calcium titanate and stearite

The loss angle of zinc titanate (see Fig. 6) decreases strongly with increasing frequency, and in the region of high frequencies (above 10^7 cps) it has a small frequency-independent value. This may be of significance for the practical use of zinc titanate in high frequency equipment.

The increase of loss angle with frequency in the region of high frequencies for all the remaining materials can be connected equally well with high frequency relaxation and with resonance absorption in the region of long infrared wavelengths. The unambiguous decision of this question requires further investigation in the shorter radio wavelength region and in the infrared region of the spectrum.

The temperature dependence of ϵ and $\tan \delta$ at frequencies of 10^3 , 10^6 , and 10^{10} cps is shown in Figs. 10-13 for the dielectrics investigated. As is clear from Figs. 10 and 11, the temperature dependence of the dielectric constant of fused strontium titanate and calcium titanate at a frequency of 10^{10} cps is completely analogous to the temperature dependence of ϵ at frequencies of 10^3 and 10^6 cps. For strontium titanate all three curves practically merge into one (Fig. 11). For calcium titanate, in

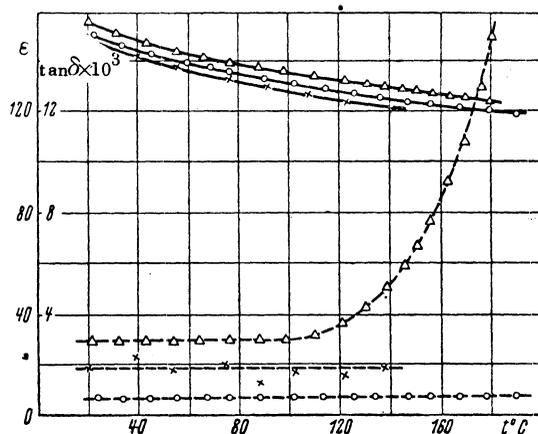


FIG. 10. Temperature dependence of ϵ and $\tan \delta$ for calcium titanate. The continuous curve gives ϵ and the dashed curve gives $\tan \delta$. Points Δ are for $f = 10^3$ cps, O for $f = 10^6$ cps, \times for $f = 10^{10}$ cps.

connection with the occurrence of some decrease in ϵ with frequency (see Fig. 8), the curves $\epsilon = f(t^\circ)$ (t is in $^\circ\text{C}$) do not coincide for different frequencies. However, they have the same slope. From this

there follows the practically important consequence that the temperature coefficient of capacity for ceramic condensers based on strontium and calcium titanates, cannot depend strongly on frequency right up to frequencies of $\sim 10^{10}$ cps. As is well known, the temperature coefficient of the dielectric constant of magnesium titanate and barium tetratitanate is comparatively small $(1/\epsilon)(d\epsilon/dT) \approx (10-40) \times 10^{-6}$ degree $^{-1}$; hence it does not seem possible to show the change in ϵ with temperature for these materials by means of the measuring apparatus used in the present work. The practical constancy of the dielectric constants of magnesium titanate and barium tetratitanate with change in temperature (Fig. 12) only emphasizes the possibility of temperature measurements by means of a resonator with a heating coil, for changes in ϵ which are not very small.

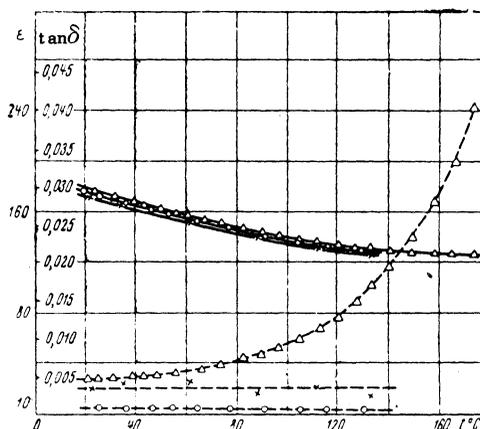


FIG. 11. Temperature dependence of ϵ and $\tan \delta$ for strontium titanate. The continuous curve gives ϵ and the dashed curve gives $\tan \delta$. Points Δ are for $f = 10^3$ cps, O for $f = 10^6$ cps, \times for $f = 10^{10}$ cps.

The increase in ϵ with temperature for bismuth titanate (Fig. 13), occurring especially at a frequency of 10^3 cps, emphasizes the presence of relaxation polarization with a comparatively large relaxation time. The essential thing is that even at high frequencies the ϵ of bismuth titanate grows with temperature. It should be noted that the bismuth titanate investigated in the present work has a complex structure, differing strongly from the structure of rutile and perovskite, and hence it is very difficult to determine to what extent its structure favors the polarization. It may clearly be asserted that in bismuth titanate there are weakly bound ions or groups of ions giving rise to relaxation (thermal ionic) polarization.

The dielectric loss angle of calcium titanate

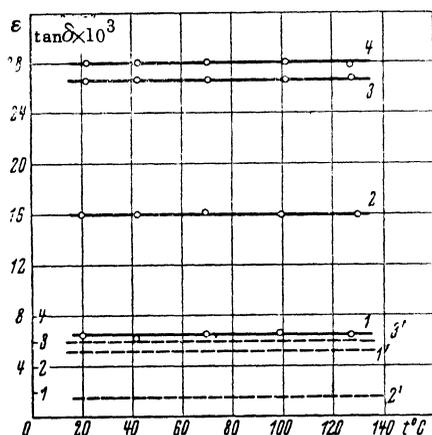


FIG. 12. Temperature dependence: The continuous curves give ϵ and the dashed curves $\tan \delta$; 1 and 1' - steatite, 2 and 2' - magnesium titanate, 3 and 3' - barium tetratitanate, 4 - zinc titanate. The points of curves 1', 2' and 3' give a scatter of (20 - 50)%.

and fused strontium titanate in the temperature region from room temperature to 160° increases strongly with temperature only at low frequencies ($f = 10^3$ cps, Figs. 10 and 11). At frequencies of 10^6 and 10^{10} cps the $\tan \delta$ of these dielectrics is constant in this temperature interval. The loss angle of magnesium titanate and barium tetratitanate at a frequency of 10^{10} cps does not change in the temperature interval 20 - 120° .

It is evident from Fig. 13 that the loss angle of bismuth titanate is independent of temperature in the interval 20 - 160° only for a frequency of 10^{10} cps. At lower frequencies (10^6 , 10^3 cps) the loss angle of bismuth titanate increases more strongly with temperature the lower the frequency. This fact is connected with the presence of relaxation polarization. The investigation of solid dielectrics in the long wavelength part of the infrared region of the spectrum will permit our conclusions concerning the nature of dielectric polarization and loss to be developed and made more precise.

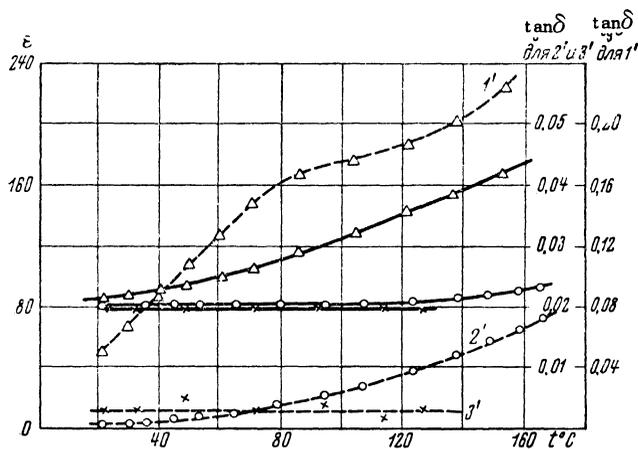


FIG. 13. Temperature dependence of ϵ and $\tan \delta$ for bismuth titanate. The continuous curve gives ϵ , the dashed curve gives $\tan \delta$. Points Δ are for $f = 10^3$ cps, O for $f = 10^6$ cps, \times for $f = 10^{10}$ cps.

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